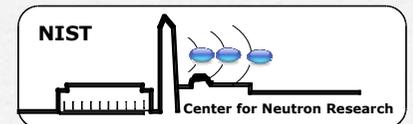


# MAGNETIC SPIN ECHO EXPERIMENTS

Jason S Gardner

NIST Center for Neutron Research  
&  
Indiana University



# Collaboration

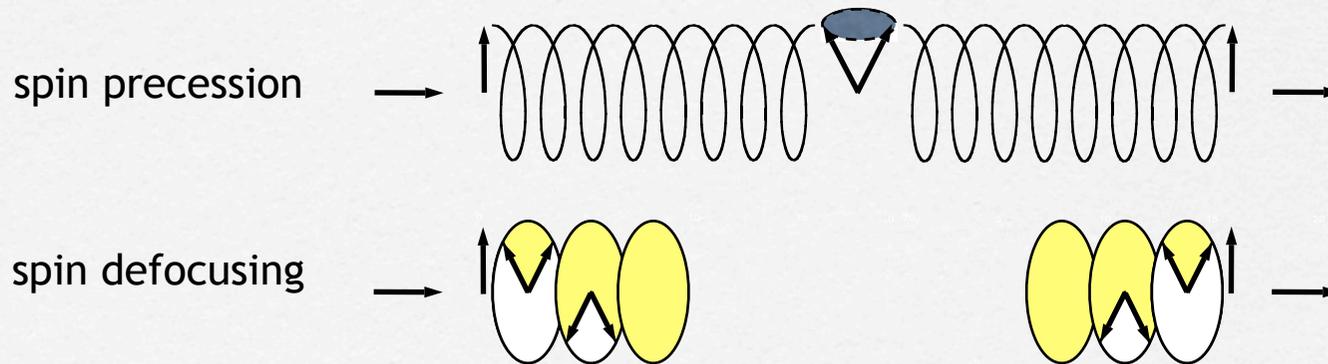
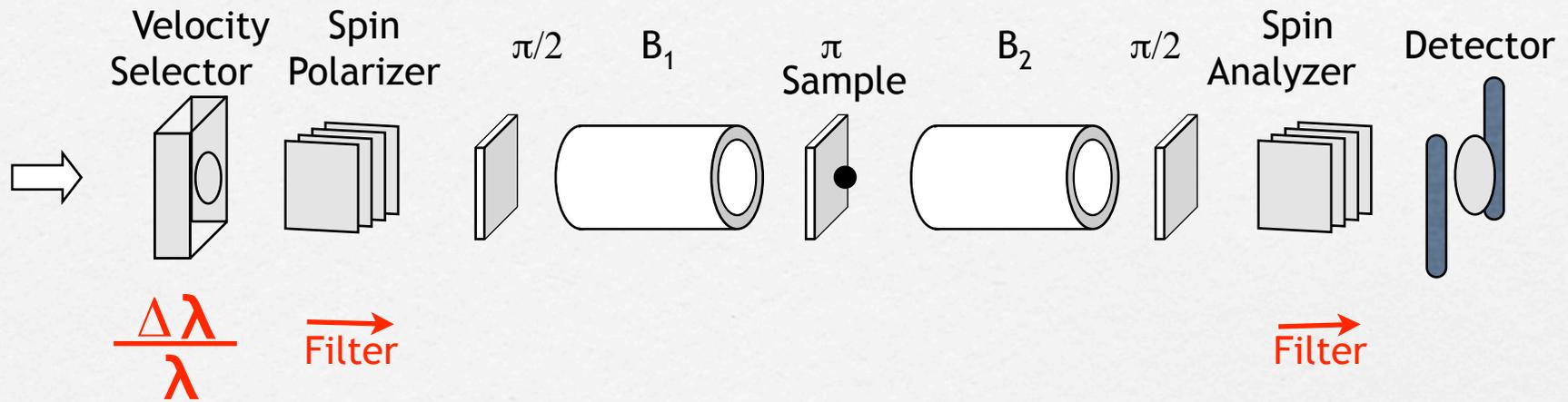
**G. Ehlers**, *SNS, Oak Ridge National Laboratory, Oak Ridge, TN*

S. T. Bramwell and T Fennell, *University College London, London WC1H  
0AJ, UK*

R. Stewart, *ILL, Grenoble, France*

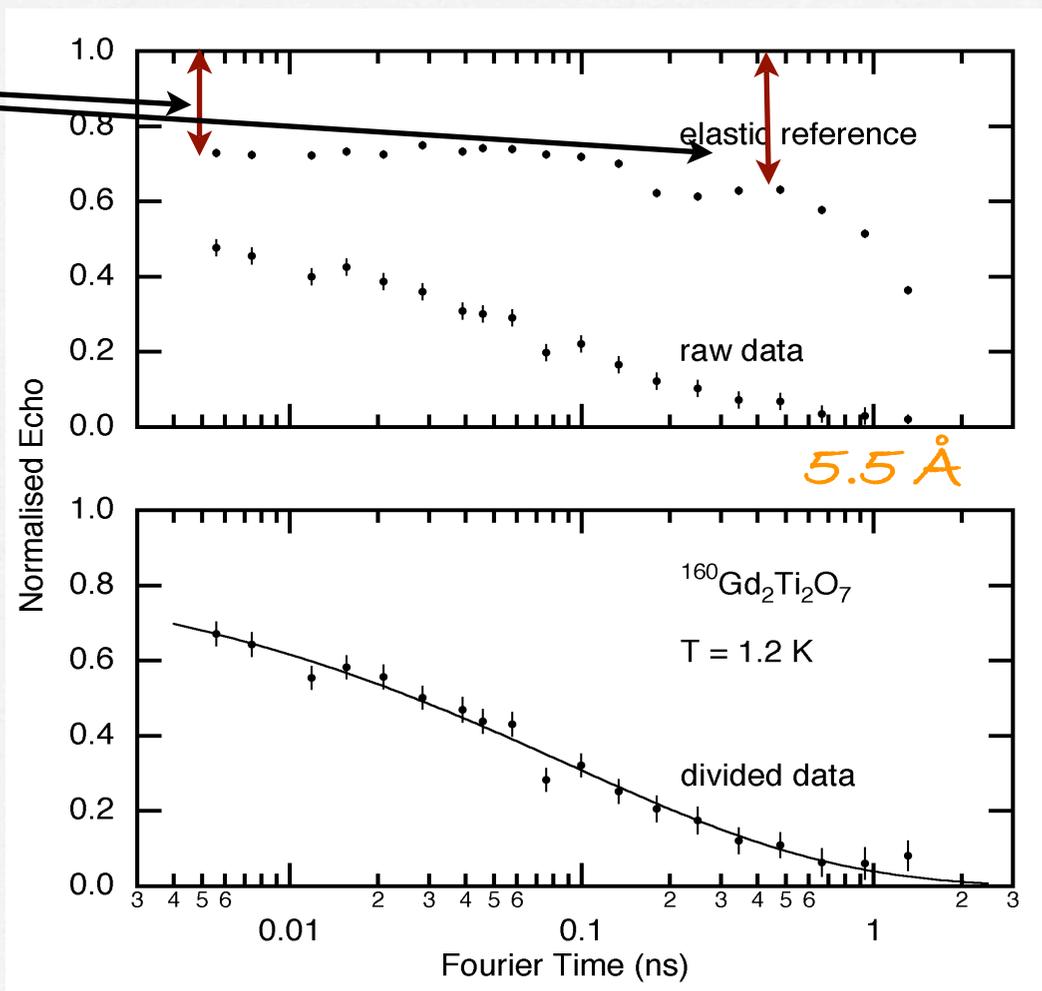
A. L. Cornelius, *University of Nevada Las Vegas, Las Vegas, NV 89154.*

# Spin Echo - How does it work



# Data Analysis Procedure

Imperfections in the instrument



5.5 Å

$\lambda^3$

# Outline



Introduction and Repetition



Peculiar characteristics of magnetic scattering:  
intensity considerations and spin flip scattering



Science examples

Spin Glass

Spin Ice

Partial Order

Spin Liquid

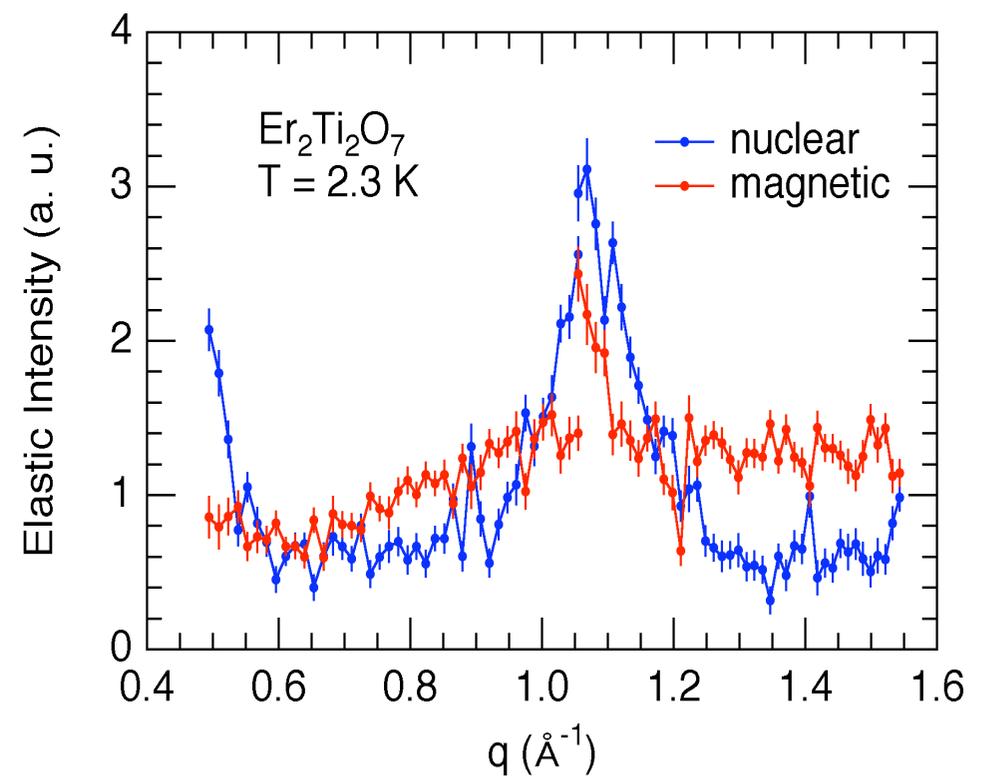
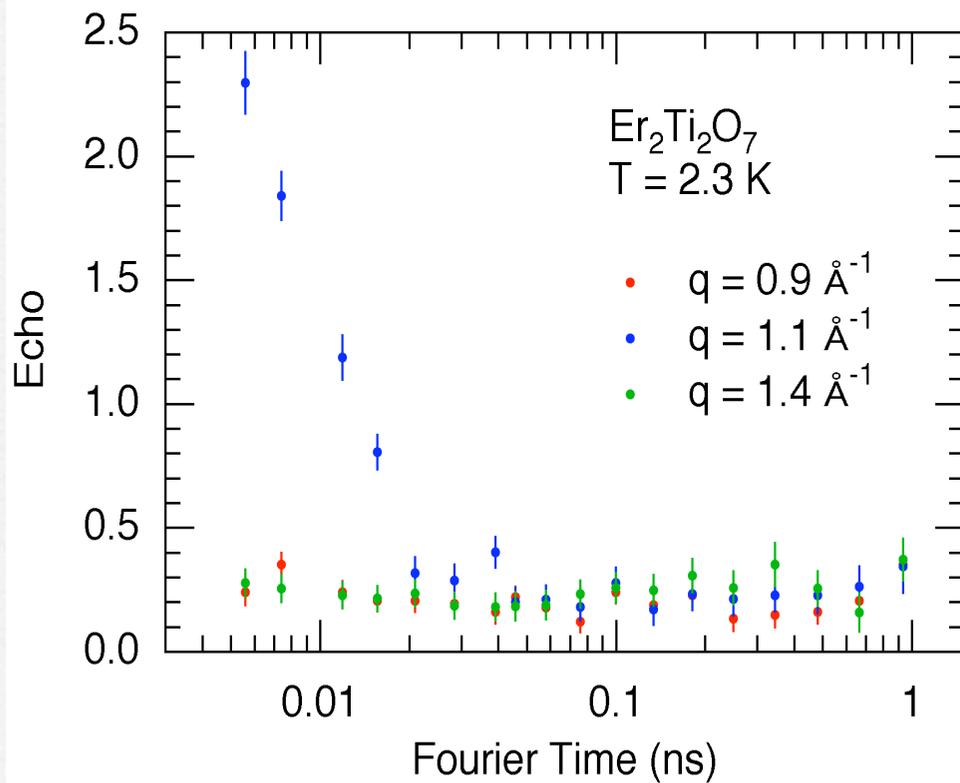


Closing remarks

# Magnetic Scattering

- The spin in the sample does the  $\pi$  flip.  
No need for the  $\pi$  flipper
- NEED xyz-analysis to scale data  
(need a resolution sample)
- Must be aware of magnetic excitations in the  
“neutron energy window” of the experiment ( $6\text{\AA}$   
 $\cong 2.3\text{meV} \Rightarrow$  Visit DCS first)
- Polarised beam so FM correlations **kill** us.
- Beware of Bragg peaks

# The NSE Spurions - Direct Echo



# xyz Polarization Analysis - the Idea

Types of scattering processes Effect on neutron spin

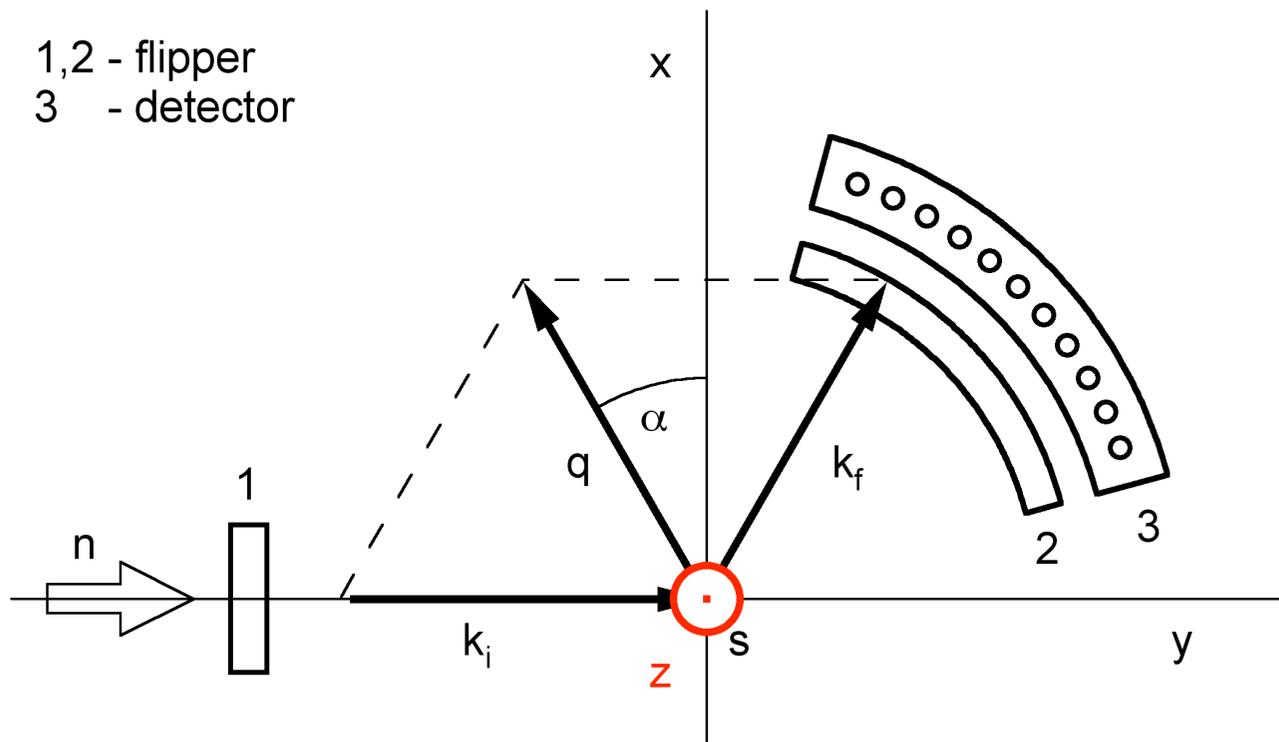
- nuclear coherent scattering (Bragg) no effect
- isotope incoherent scattering no effect
- spin incoherent scattering spin flip (2/3 probability)
  - magnetic scattering spin rotation

	$B$ vertical	$B$ horizontal ( $\perp \mathbf{q}$ )	$B$ horizontal ( $\parallel \mathbf{q}$ )
$\mu$ vertical	<i>nsf</i>	<i>sf</i>	<i>sf</i>
$\mu$ horizontal ( $\perp \mathbf{q}$ )	<i>sf</i>	<i>nsf</i>	<i>sf</i>
$\mu$ horizontal ( $\parallel \mathbf{q}$ )	0	0	0
average	50% <i>nsf</i> 50% <i>sf</i>	50% <i>nsf</i> 50% <i>sf</i>	100% <i>sf</i>

in a paramagnet,  
magnetic moment  
directions are  
isotropically  
distributed

# xyz Polarization Analysis - Reality

1,2 - flipper  
3 - detector



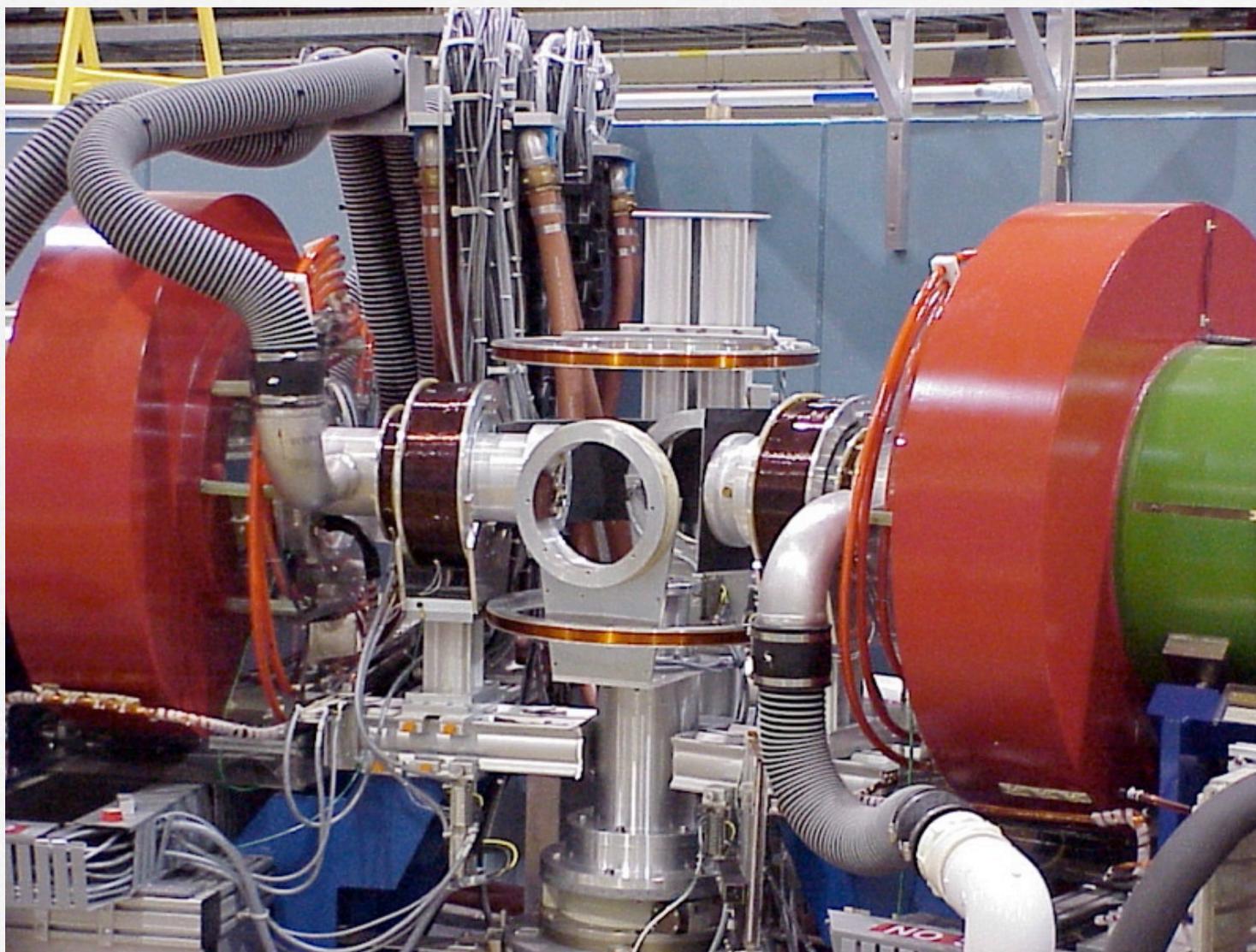
# xyz Polarization Analysis - Equations

case	non spin flip ("up")	spin flip ("down")
$\vec{p} \parallel \vec{x}$	$\sigma_n \frac{1+p}{2} + \frac{1}{2} \sigma_m \cdot (1 - p \cdot \cos^2 \alpha)$	$\sigma_n \frac{1-p}{2} + \frac{1}{2} \sigma_m \cdot (1 + p \cdot \cos^2 \alpha)$
$\vec{p} \parallel \vec{y}$	$\sigma_n \frac{1+p}{2} + \frac{1}{2} \sigma_m \cdot (1 - p \cdot \sin^2 \alpha)$	$\sigma_n \frac{1-p}{2} + \frac{1}{2} \sigma_m \cdot (1 + p \cdot \sin^2 \alpha)$
$\vec{p} \parallel \vec{z}$	$\sigma_n \frac{1+p}{2} + \frac{1}{2} \sigma_m$	$\sigma_n \frac{1-p}{2} + \frac{1}{2} \sigma_m$

$$\sigma_m = \frac{2}{p} \cdot \left( -\sigma_x^{\text{up}} - \sigma_y^{\text{up}} + 2 \cdot \sigma_z^{\text{up}} \right)$$

$$\sigma_m = \frac{2}{p} \cdot \left( \sigma_x^{\text{down}} + \sigma_y^{\text{down}} - 2 \cdot \sigma_z^{\text{down}} \right)$$

# NG5NSE



# Outline



Introduction and Repetition



Peculiar characteristics of magnetic scattering:  
intensity considerations and spin flip scattering



## Science examples

Spin Glass

Spin Ice

Partial Order

Spin Liquid



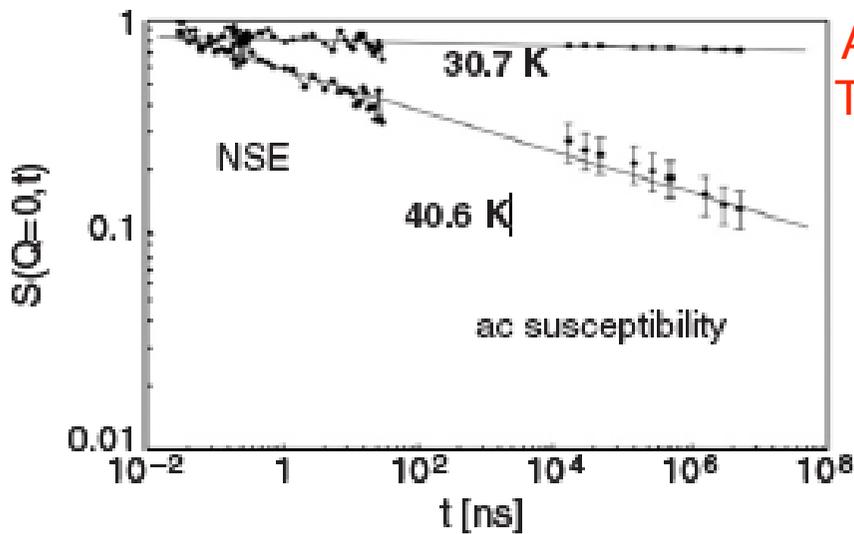
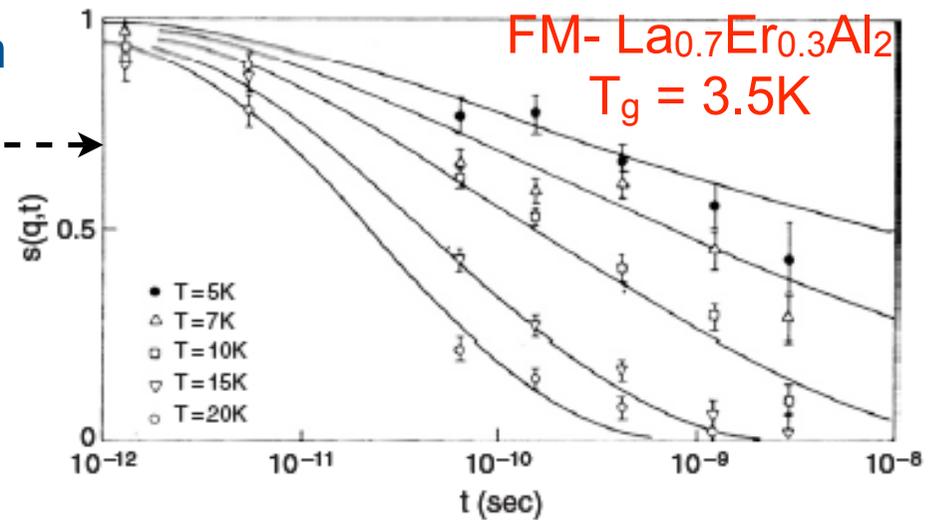
Closing remarks

# SPIN GLASSES

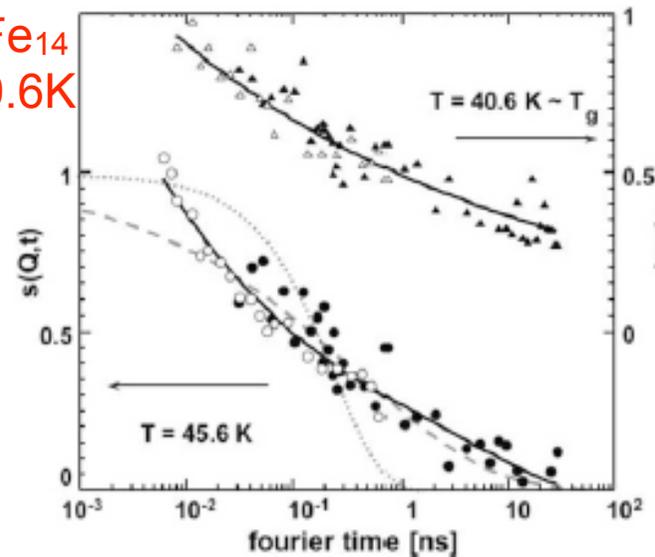
Model assuming different  $\tau$  and  $E$  in different parts of the sample.  
 Mezei, Solid State Comm. 45 411, 1983

At  $T_g$  and below - power law  
 above  $T_g$  - Ogielski function  
 B increases from 1/3

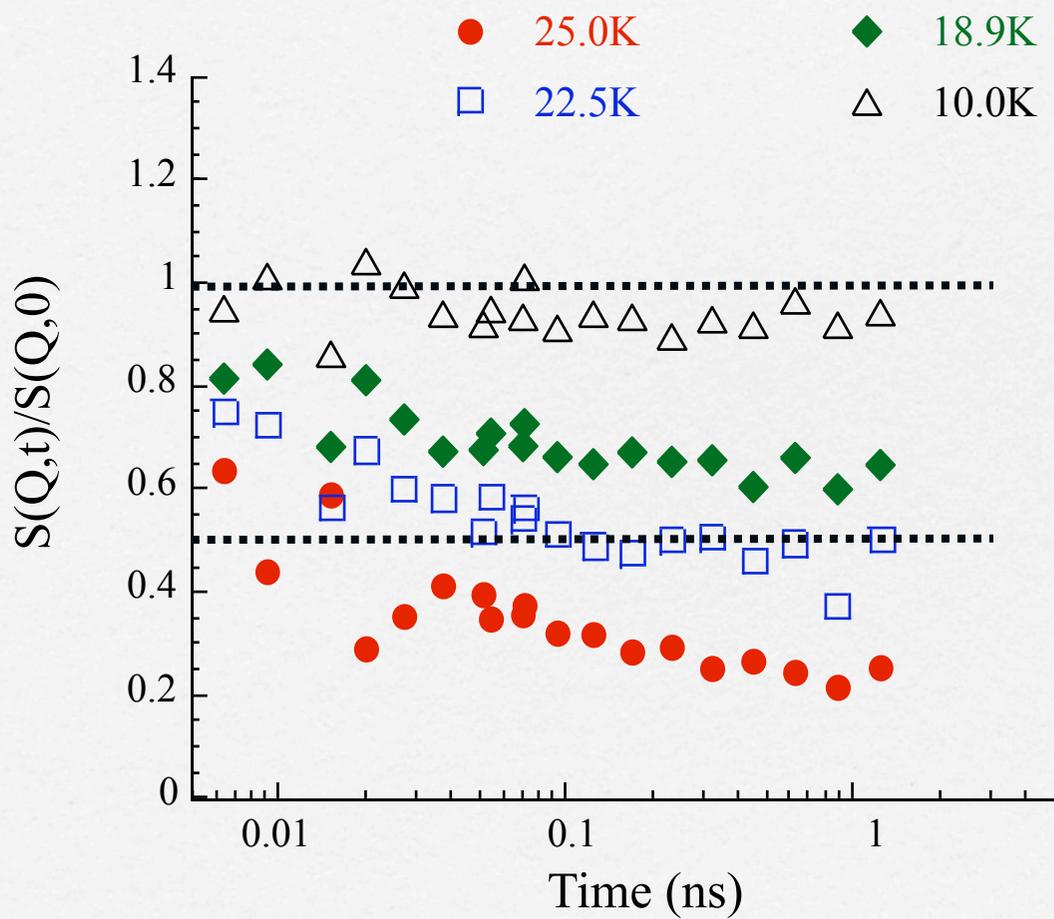
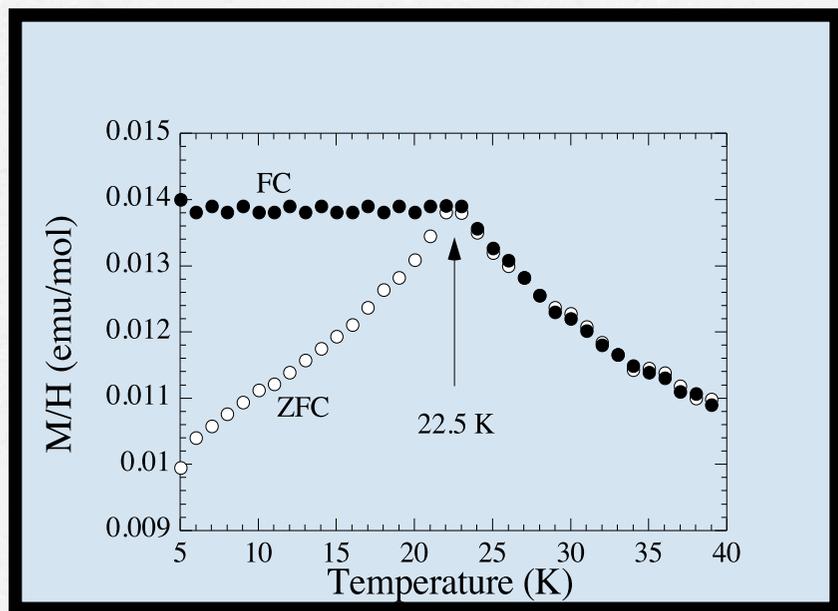
Pappas, Solid State Comm. 68 054431, 2003



$\text{Au}_{86}\text{Fe}_{14}$   
 $T_g = 40.6\text{K}$



# SPIN GLASSES - $Y_2MgO_7$



# Outline



Introduction and Repetition



Peculiar characteristics of magnetic scattering:  
intensity considerations and spin flip scattering



## Science examples

Spin Glass

Spin Ice

Partial Order

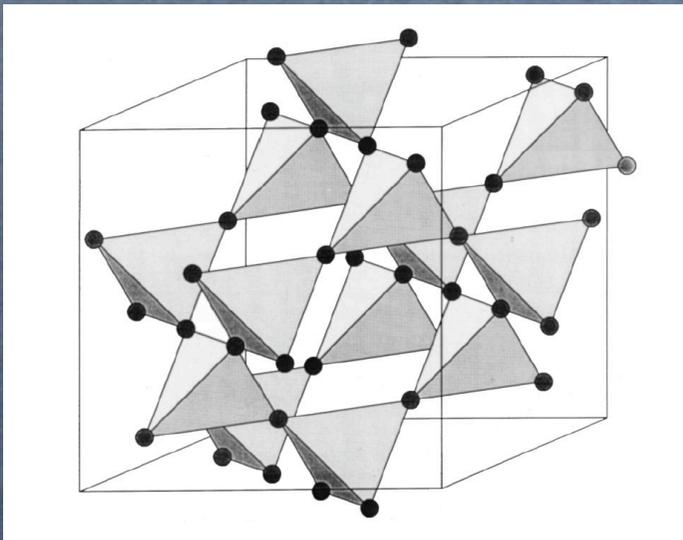
Spin Liquid



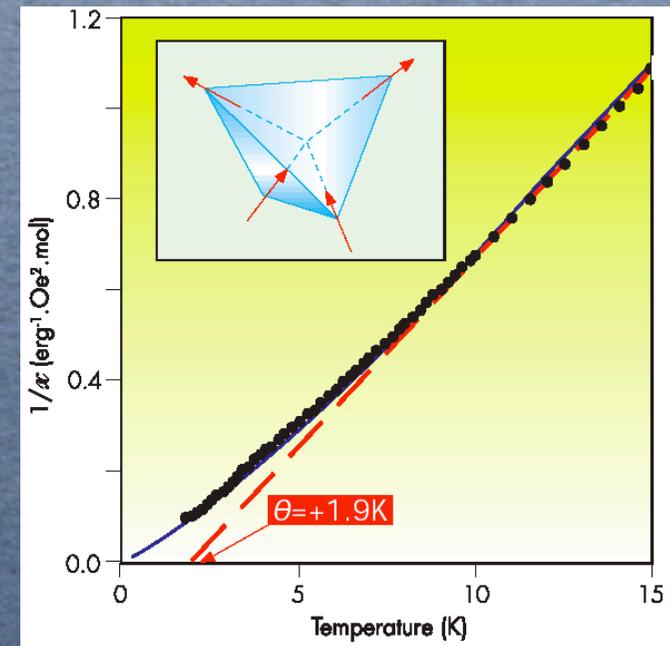
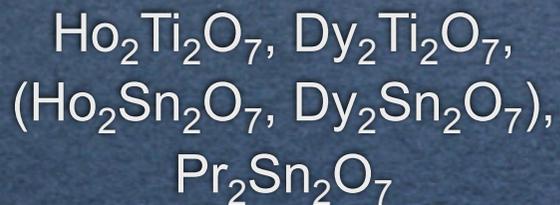
Closing remarks

# SPIN ICE

Spin Ice Crystal Structure



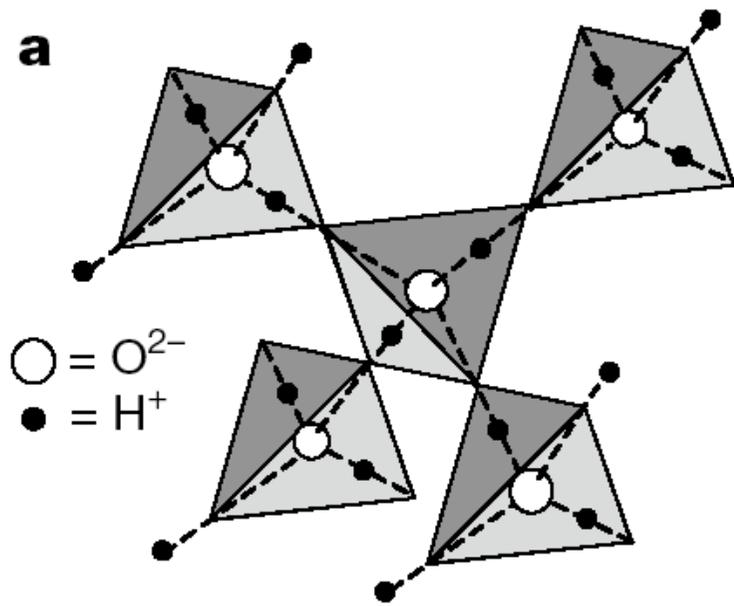
Stoichiometry



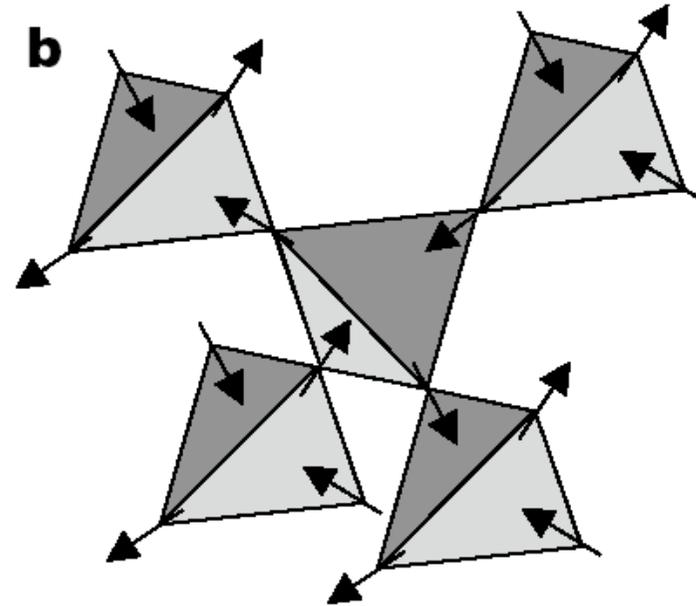
$$\chi(T) = \frac{n}{3k_B} \cdot \frac{\mu_{\text{eff}}^2}{T - \Theta}$$

$$\mu_{\text{eff}} \approx 9\mu_B \quad \Theta \approx +1\text{K}$$

# ANALOGY TO WATER ICE



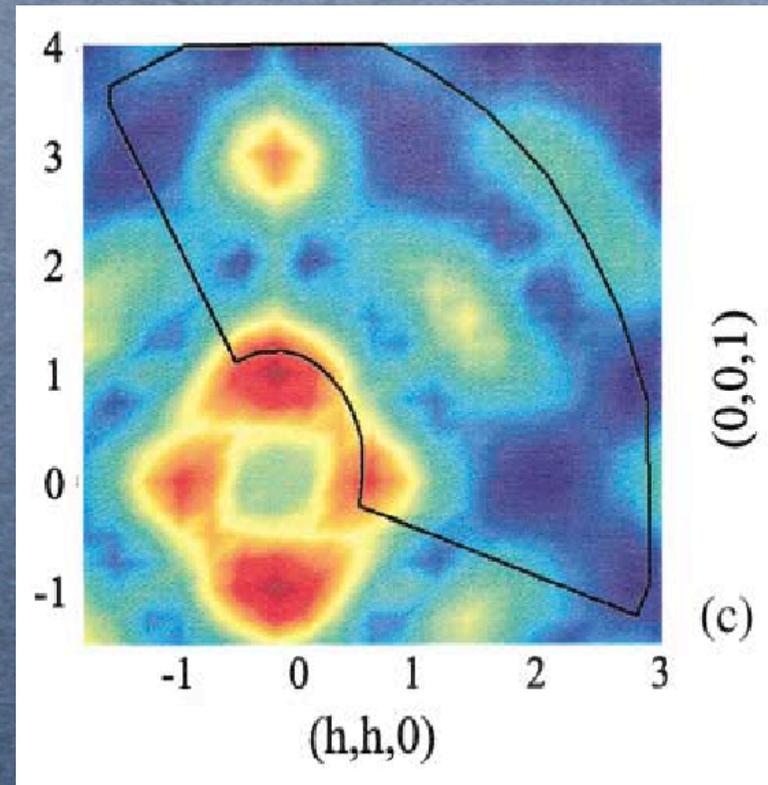
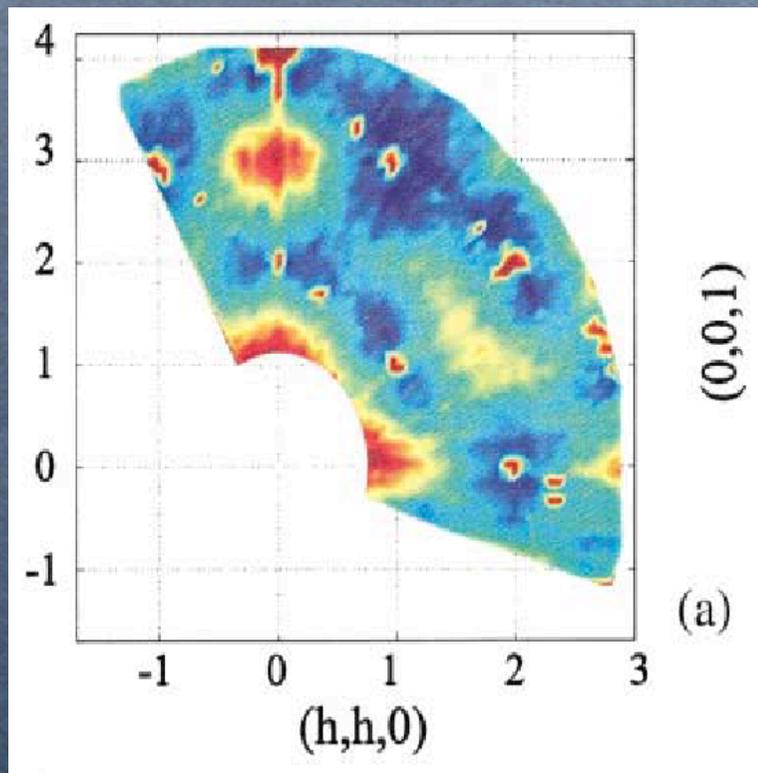
Water ice



Spin ice

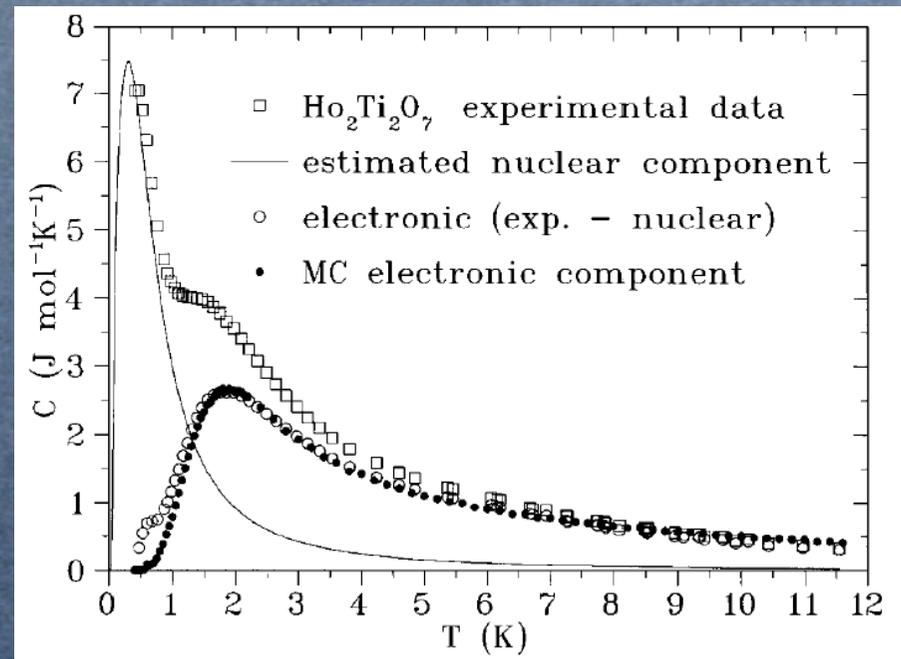
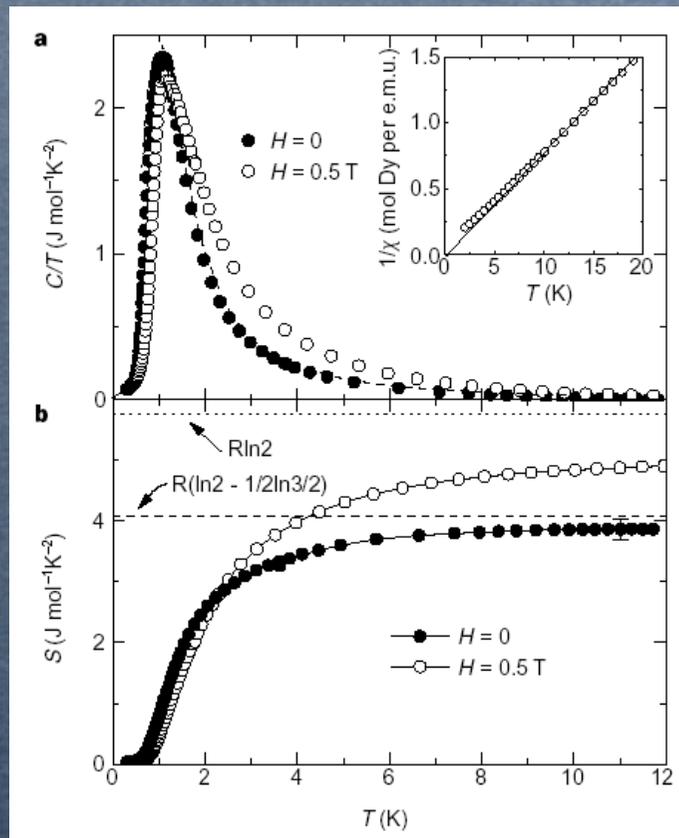
CEF constraints spins to local  $\langle 111 \rangle$  axes

# NEUTRON DIFFRACTION



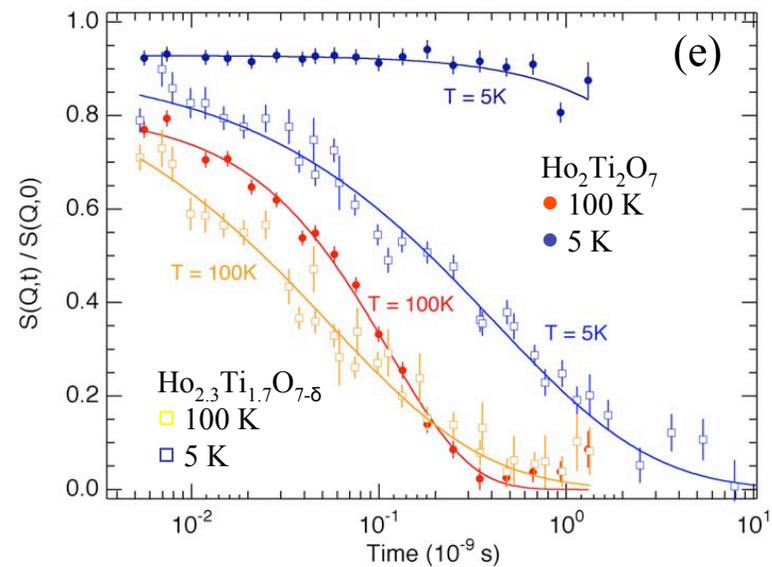
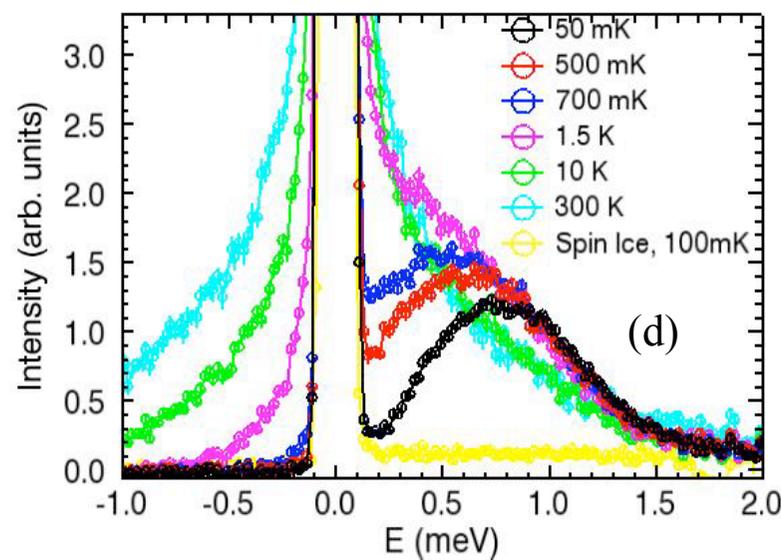
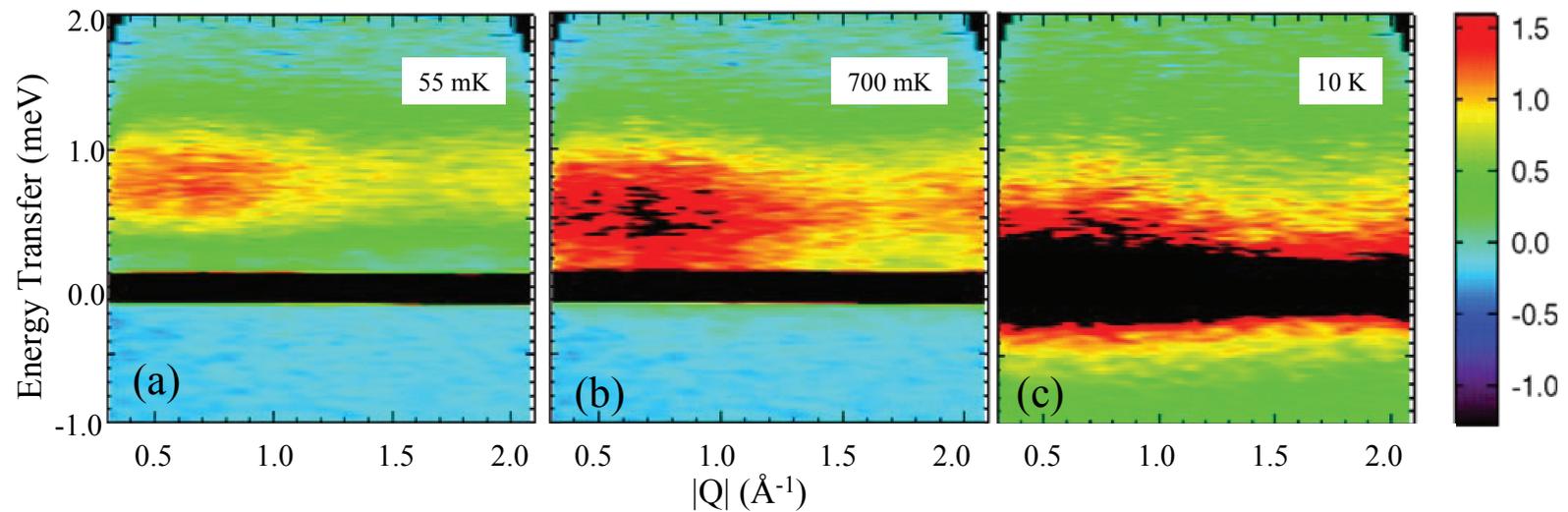
PRISMA  $T = 50$  mK S. T. Bramwell et al.,  
PRL 87, 047205 (2001)

# MAGNETIC SPECIFIC HEAT

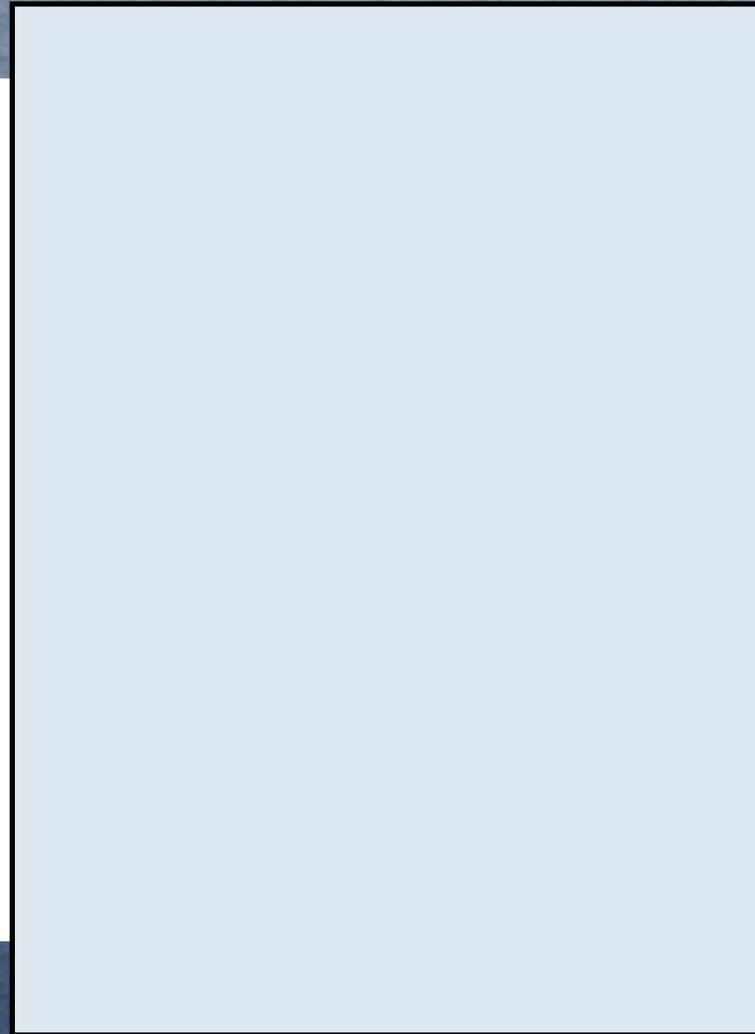
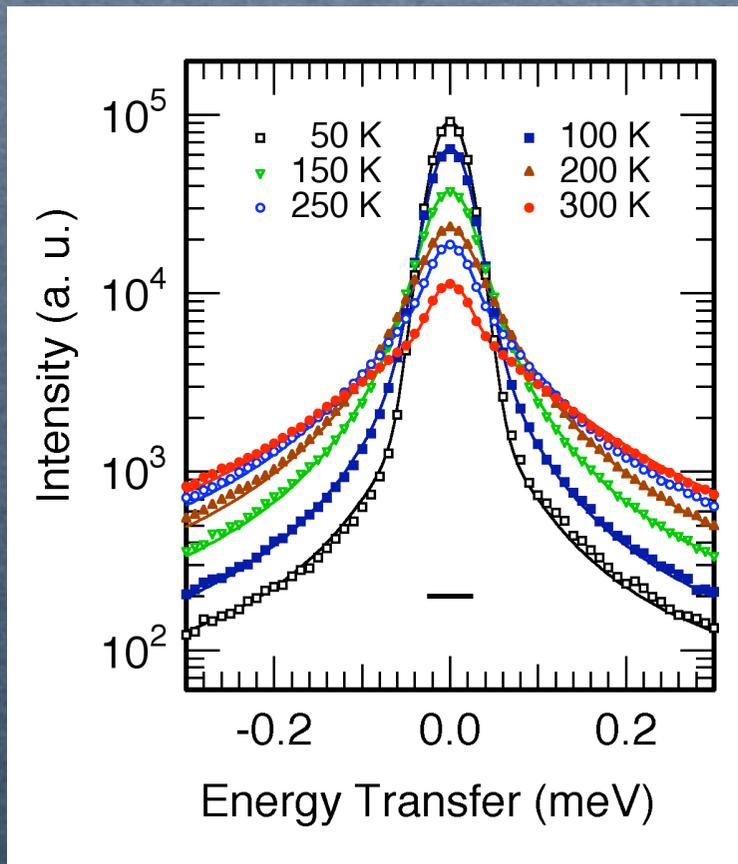


A. P. Ramirez et al., S. T. Bramwell et al.,  
Nature 399, 333 (1999). PRL 87, 047205 (2001).

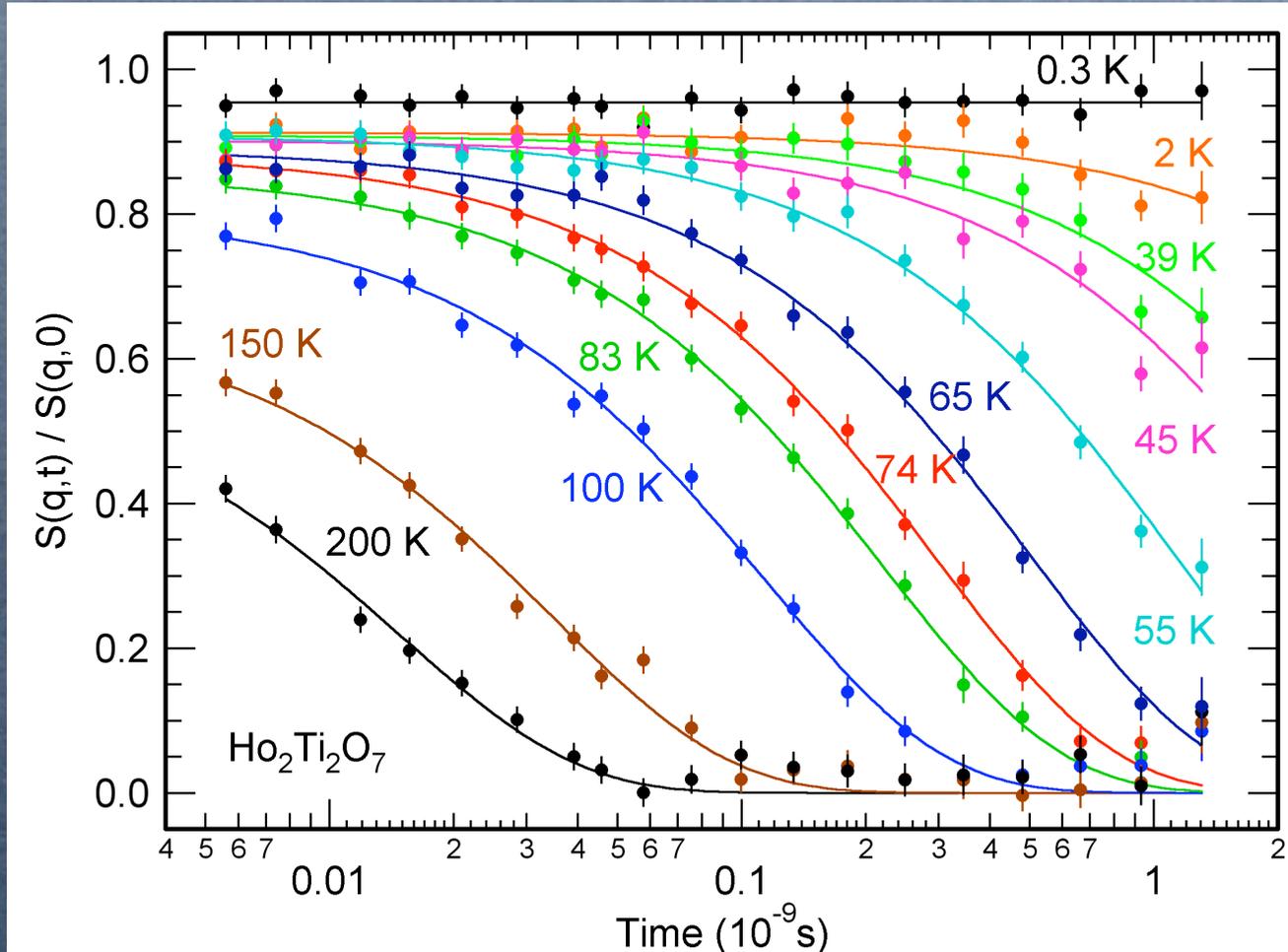
# PREVIEW



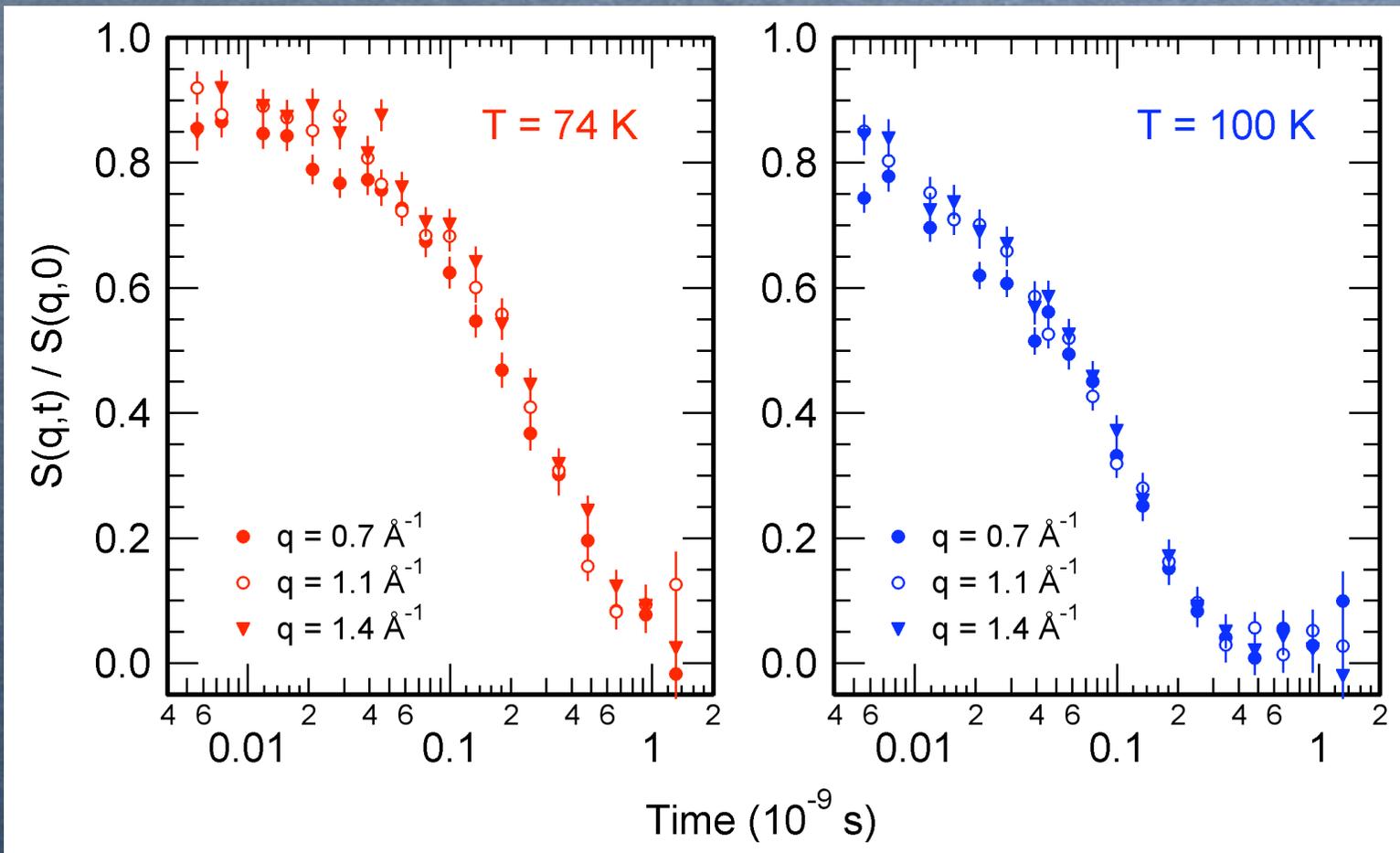
# MORE INELASTIC RESULTS



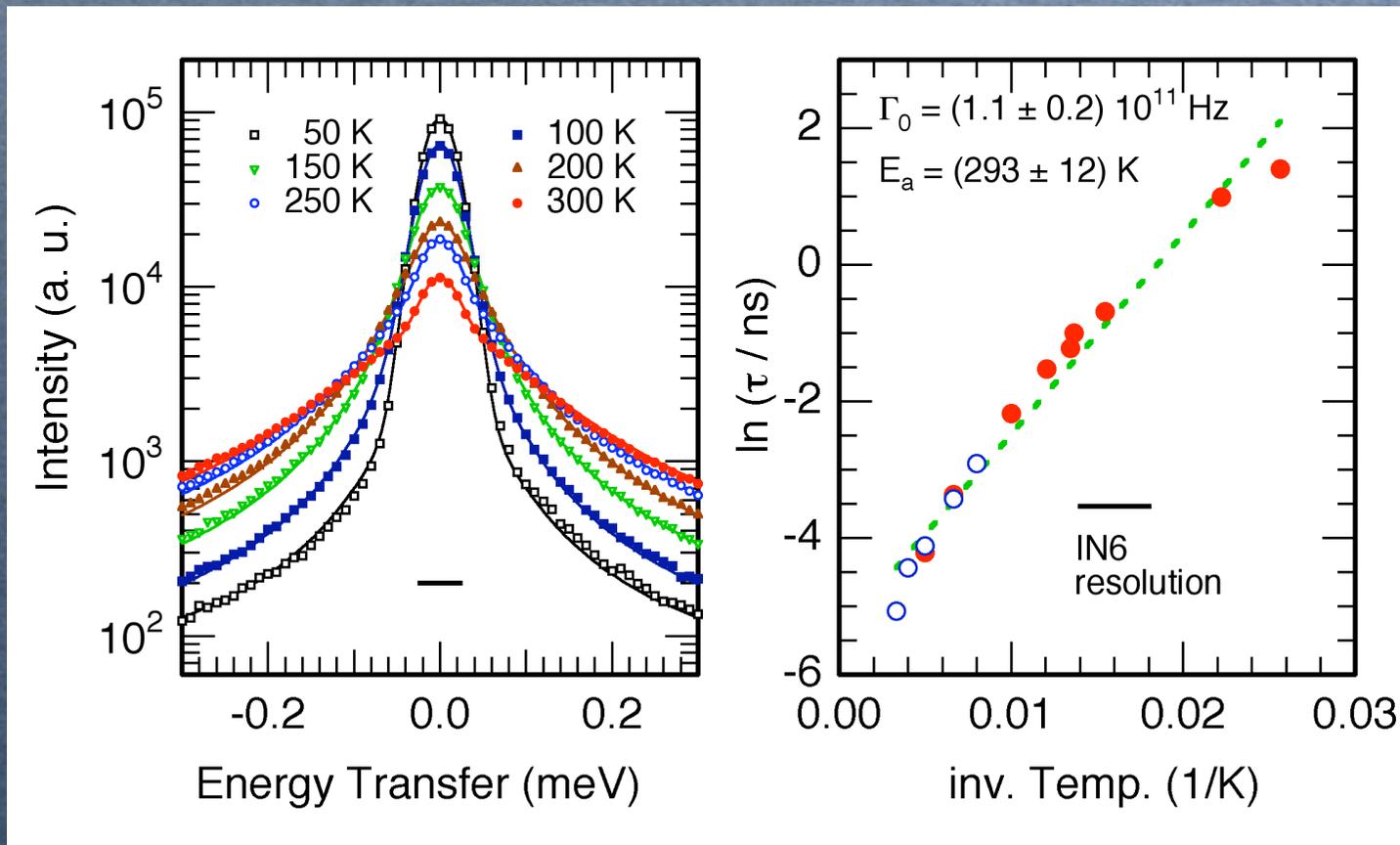
# SPIN ECHO RESULTS (1)



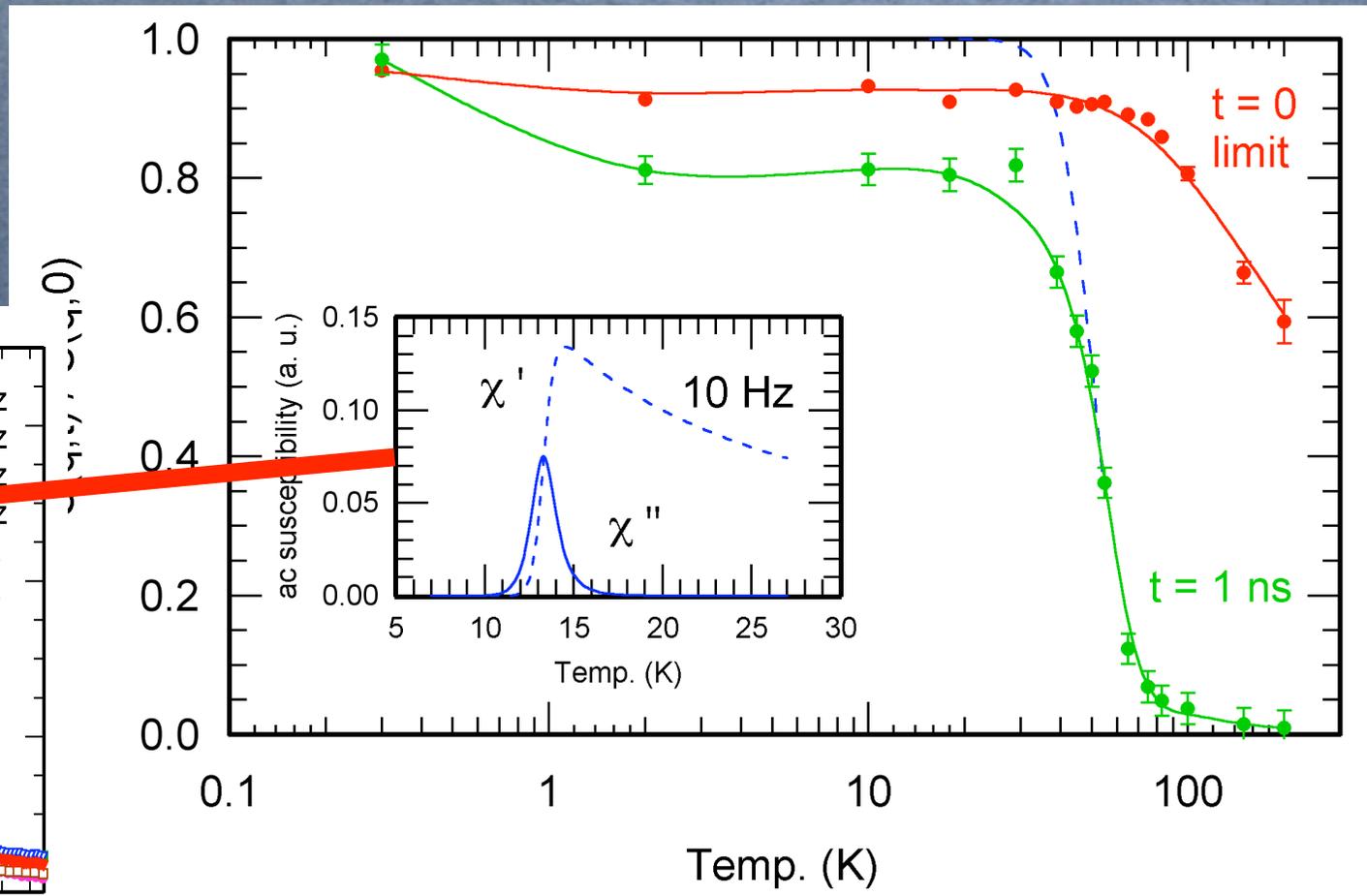
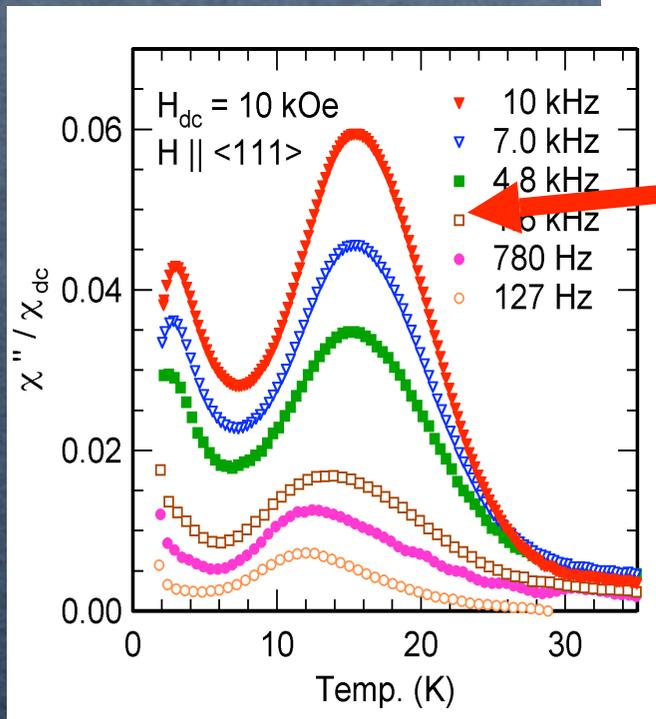
# SPIN ECHO RESULTS (2)



# MORE INELASTIC RESULTS



# COMPARISON WITH AC SUSCEPTIBILITY



# CONCLUSION

NSE works perfectly to study the dynamics of spin ice:

- works in the time domain
- gives just the right time range
- q information tells it is single ion relaxation
- data is 'clean' because magnetic scattering is separated

NSE proves that there are **TWO** relaxation mechanisms in spin ice.

Consistent picture with ac susceptibility results.

# Outline



Introduction and Repetition



Peculiar characteristics of magnetic scattering:  
intensity considerations and spin flip scattering



Science examples



Spin Glass



Spin Ice



Partial Order

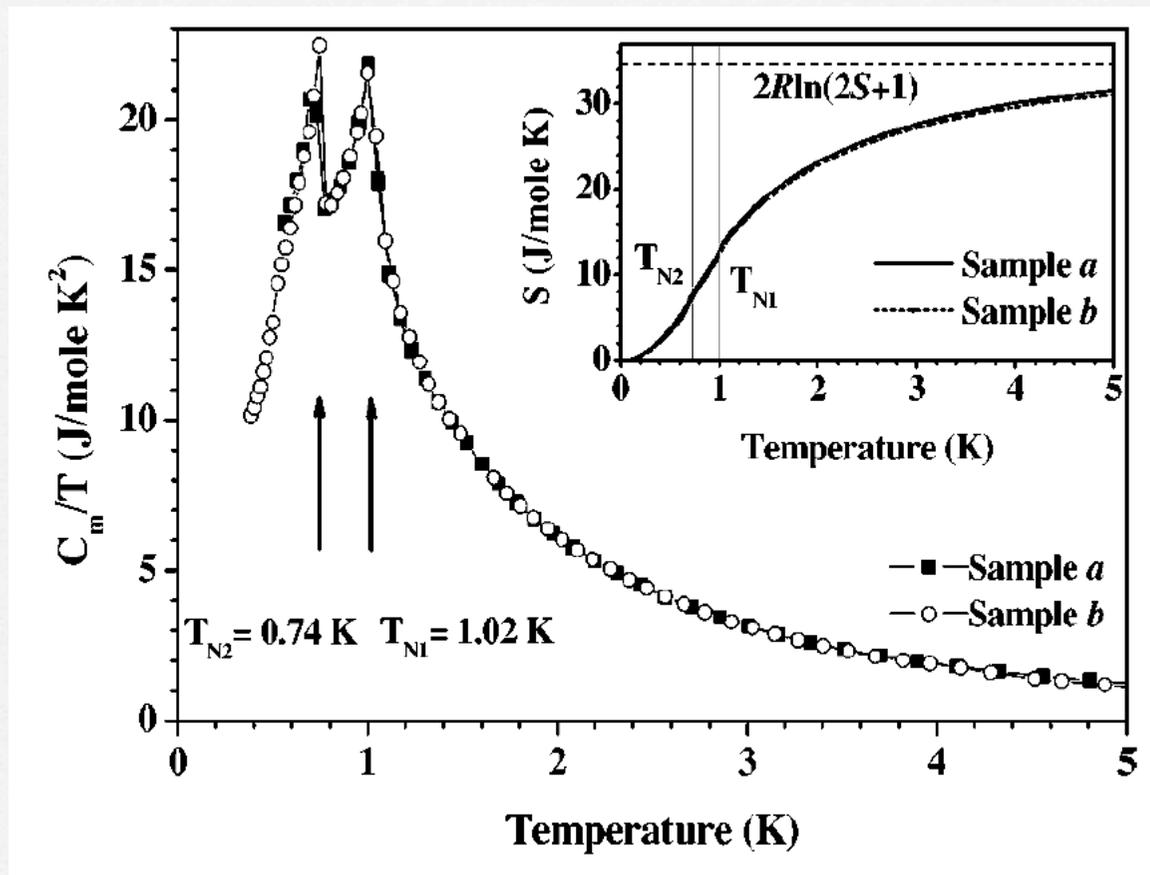


Spin Liquid



Closing remarks

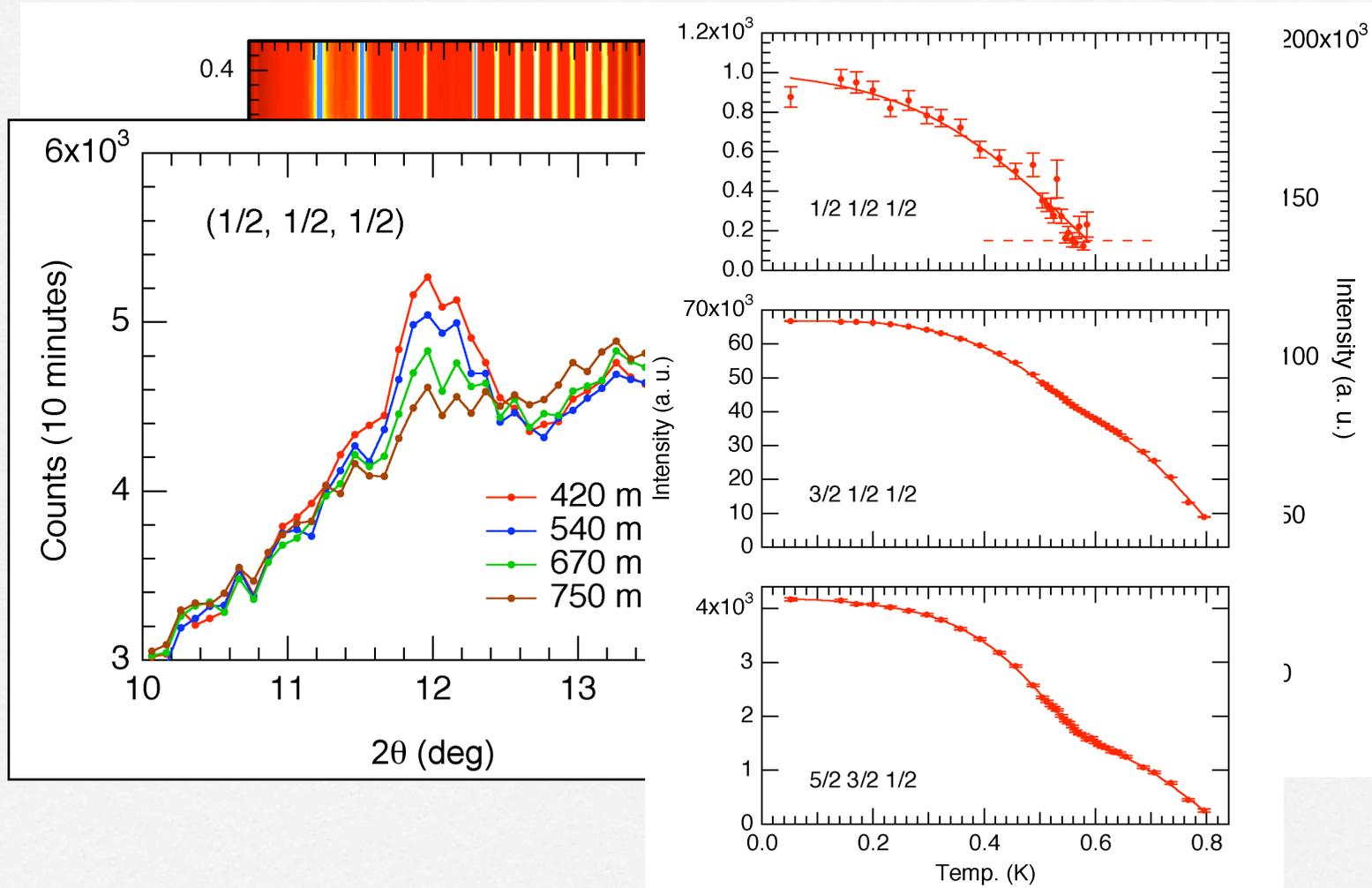
# Gd<sub>2</sub>Ti<sub>2</sub>O<sub>7</sub>



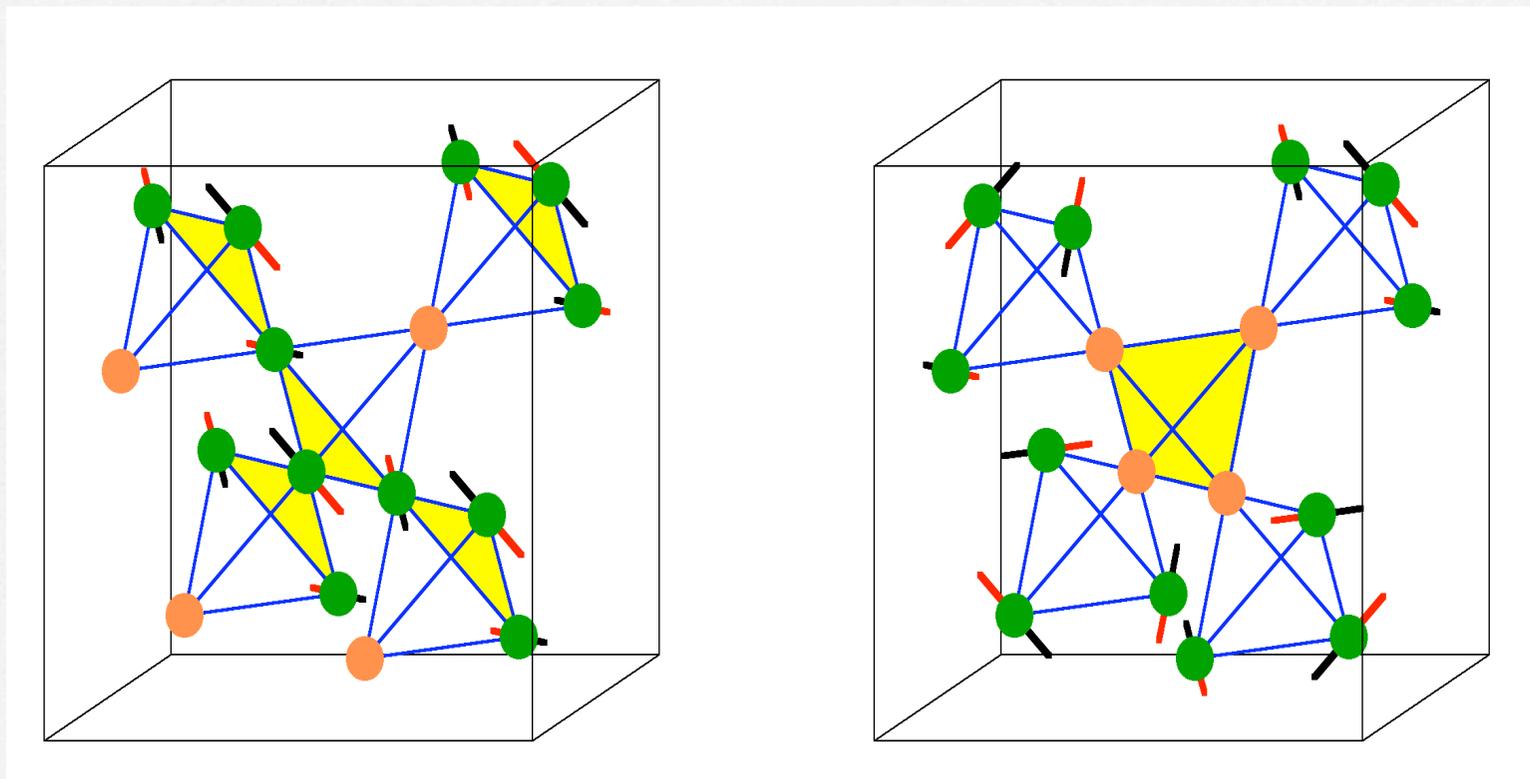
$$S = 7/2 \quad L = 0$$
$$\mu_{\text{eff}} \approx 7\mu_B \quad \Theta \approx -10\text{K}$$

O. A. Petrenko et al., PRB 70 (2004) 012402  
A. P. Ramirez et al., PRL 89 (2002) 067202

# Gd<sub>2</sub>Ti<sub>2</sub>O<sub>7</sub> - Unpolarised diffraction

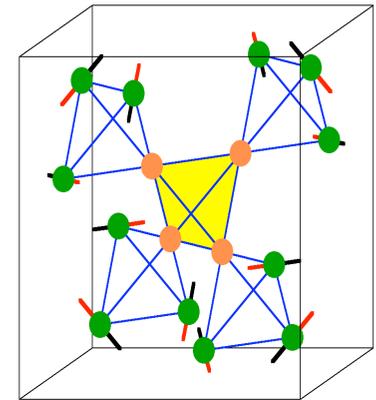
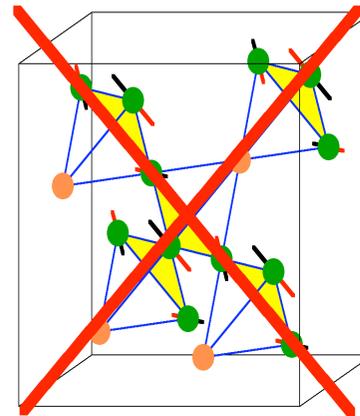
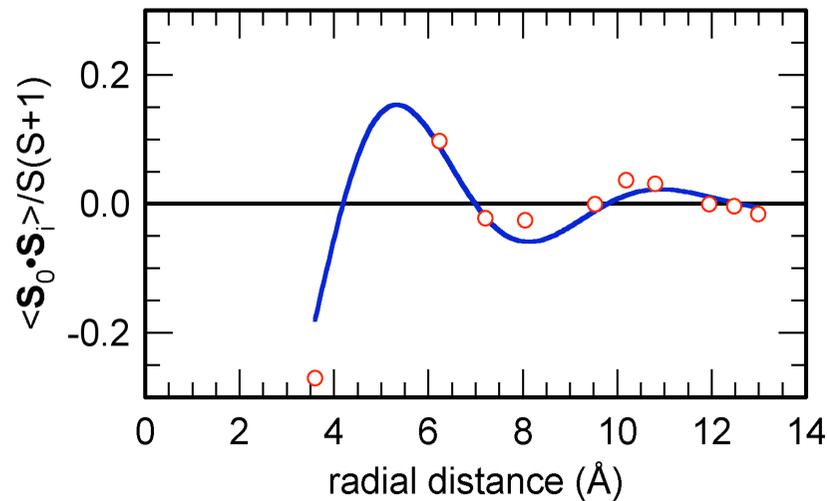
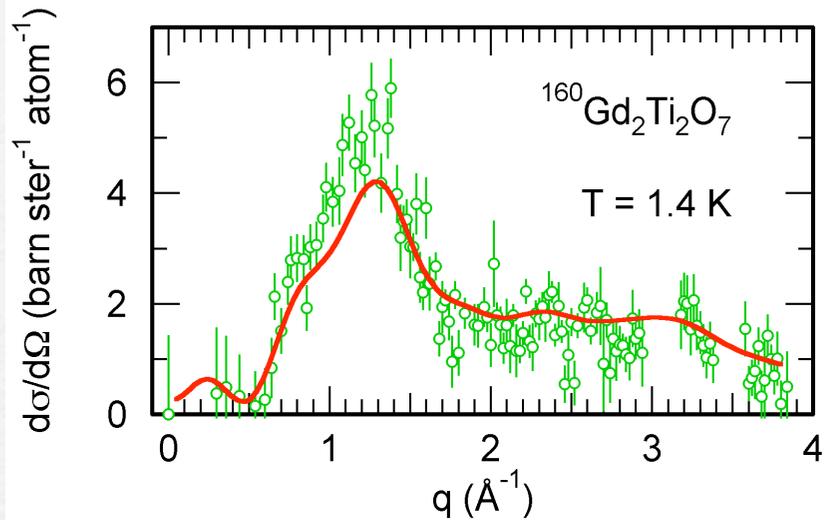


# Gd<sub>2</sub>Ti<sub>2</sub>O<sub>7</sub> - possible structures



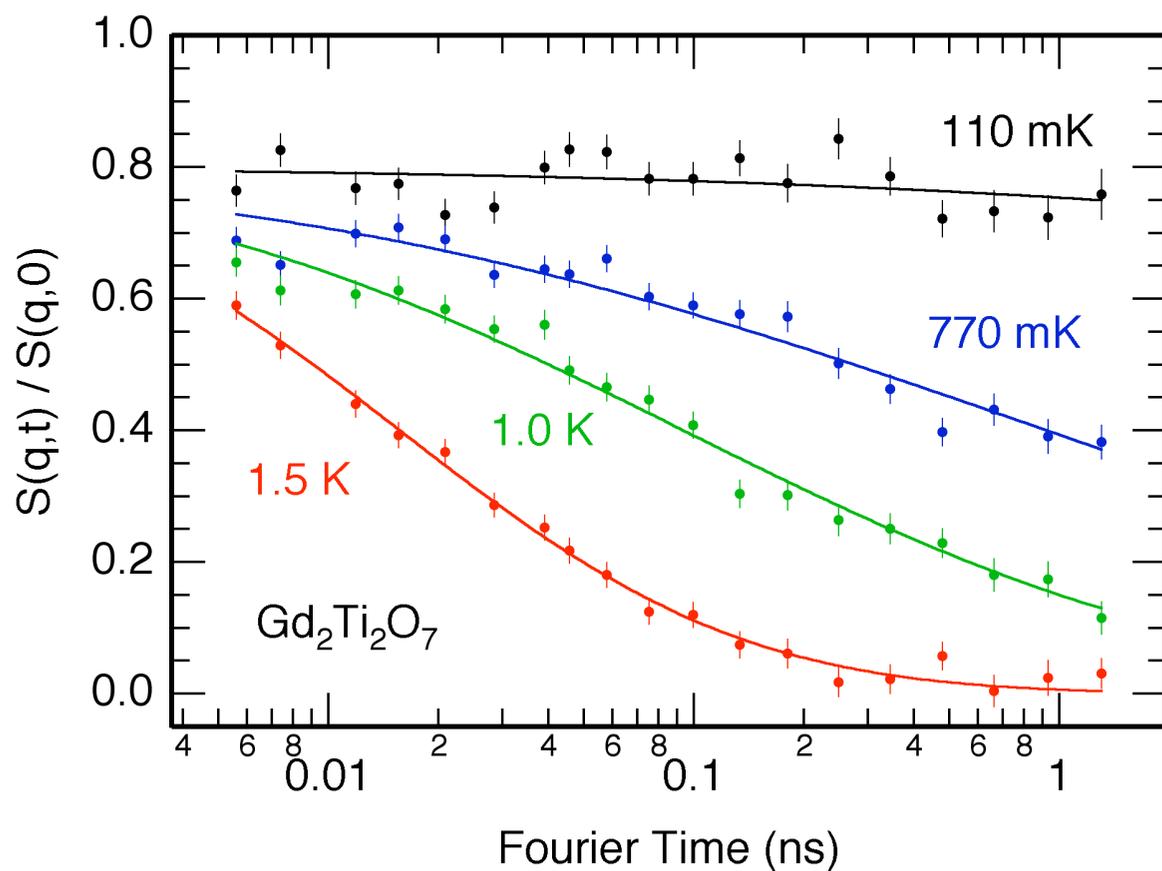
1k structure    4k structure

# Gd<sub>2</sub>Ti<sub>2</sub>O<sub>7</sub> - polarised neutrons



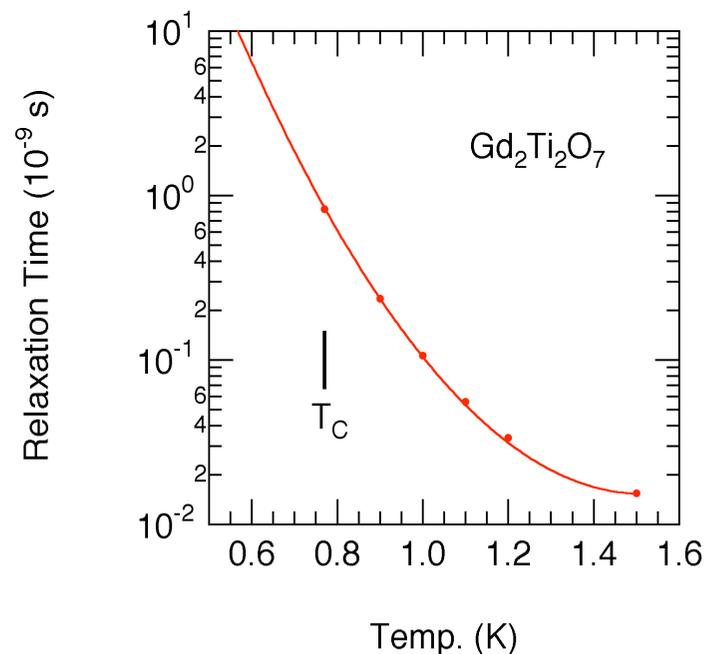
Data analysis using  
Reverse Monte Carlo  
Simulations

# Gd<sub>2</sub>Ti<sub>2</sub>O<sub>7</sub> - NSE results



$I(Q,0) \Rightarrow 0.75$  above LT transition

As the last spin begins to freeze  $I(Q,t)$  increases



## Gd<sub>2</sub>Ti<sub>2</sub>O<sub>7</sub> - NSE results

- Even small samples can be measured.
  - Rem. 1/2 g sample, 7 Bohr Magnetons
- Spin Echo reveals 3/4 spins relax with a single time constant
- With more data could reveal interesting temperature dependence.

# Outline



Introduction and Repetition



Peculiar characteristics of magnetic scattering:  
intensity considerations and spin flip scattering



## Science examples



Spin Glass



Spin Ice



Partial Order



Spin Liquid



Closing remarks

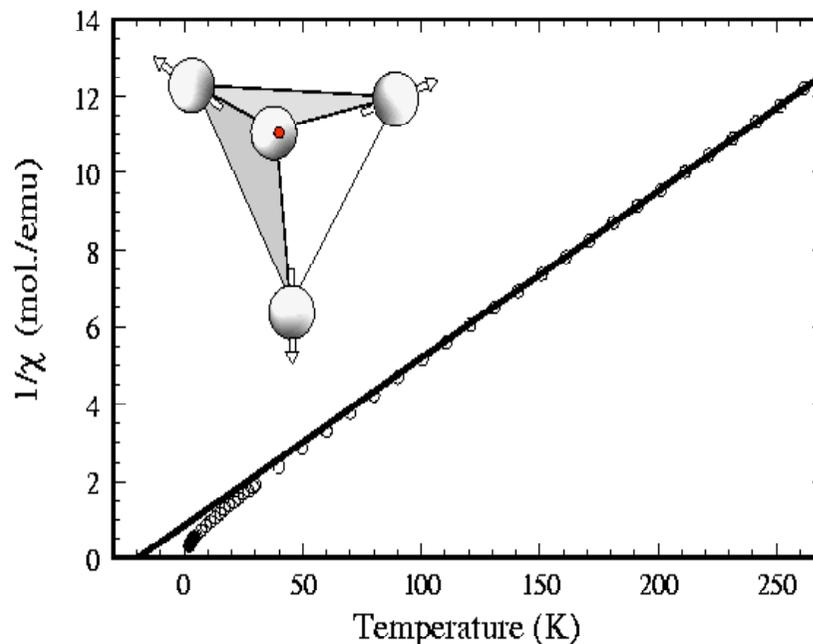
# Tb<sub>2</sub>Ti<sub>2</sub>O<sub>7</sub> - Cooperative Paramagnet

Spatial magnetic correlations of ~  
5 Å at 2 K

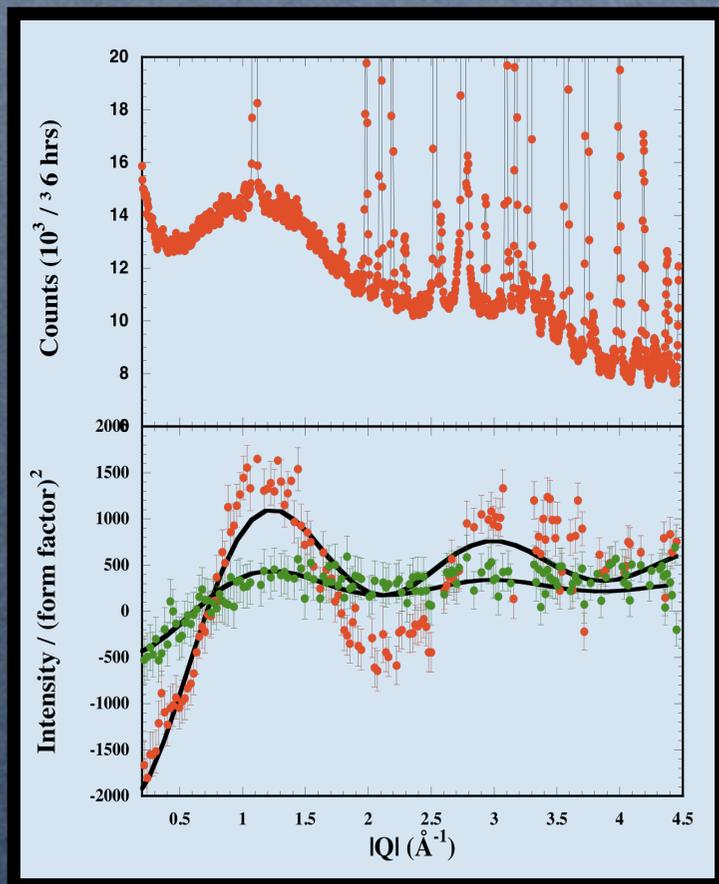
Although there are magnetic correlations below 60 K, there is **no LRO above 0.5 K.**

Mouns tell us its dynamic at 15 mK

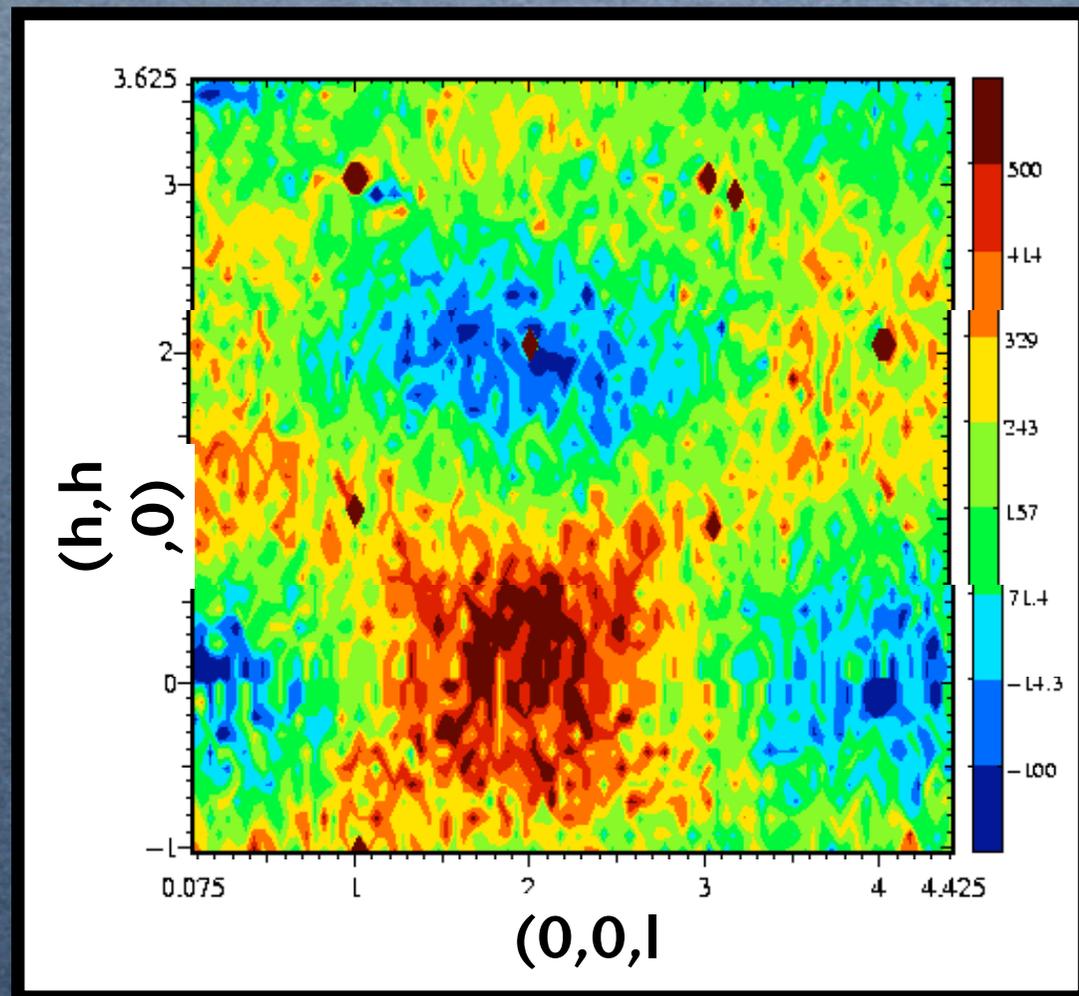
- $\Theta_{CW} = -19$  K
- $9.4 \mu_B$  ( $\sim 7F_6$  Tb<sup>3+</sup> ion)
- Very linear above  $\Theta_{CW}$
- Dips below CW behaviour



# Tb<sub>2</sub>Ti<sub>2</sub>O<sub>7</sub> - Diffraction

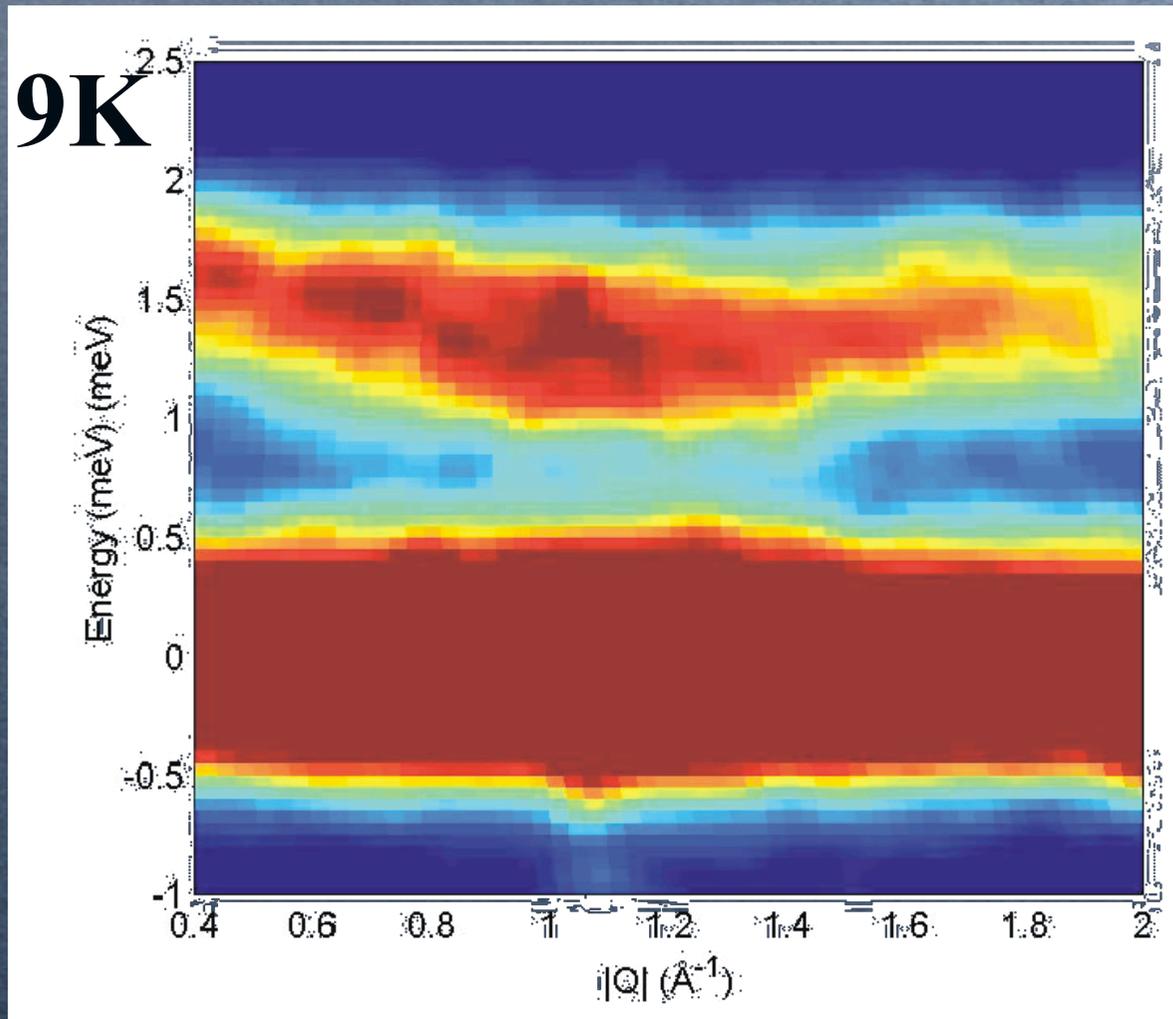


2 K



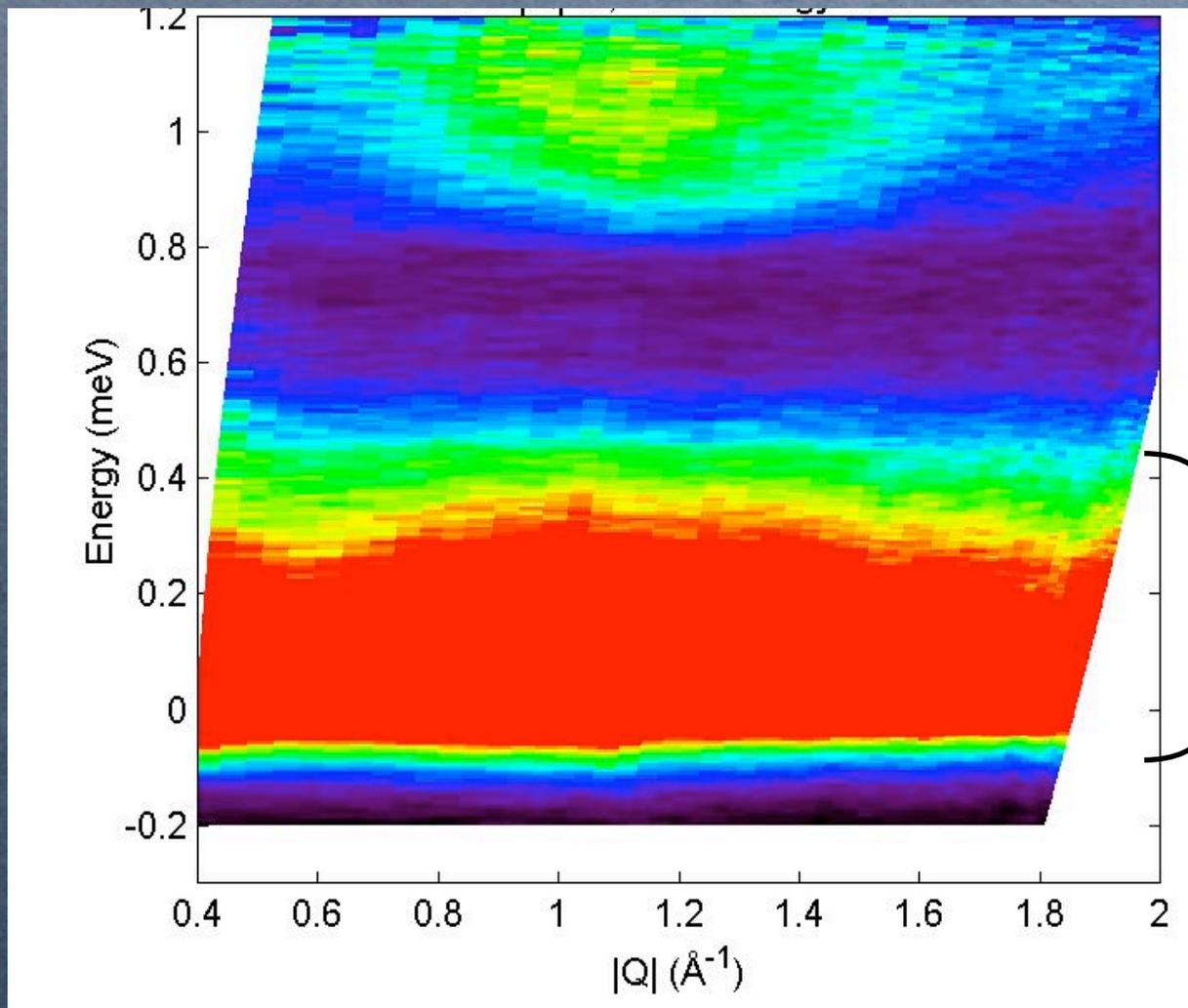
9 K

# Tb<sub>2</sub>Ti<sub>2</sub>O<sub>7</sub> - Inelastic Scattering



} Still gapped, and ...  
lots of structure

# Tb<sub>2</sub>Ti<sub>2</sub>O<sub>7</sub> - Inelastic Scattering

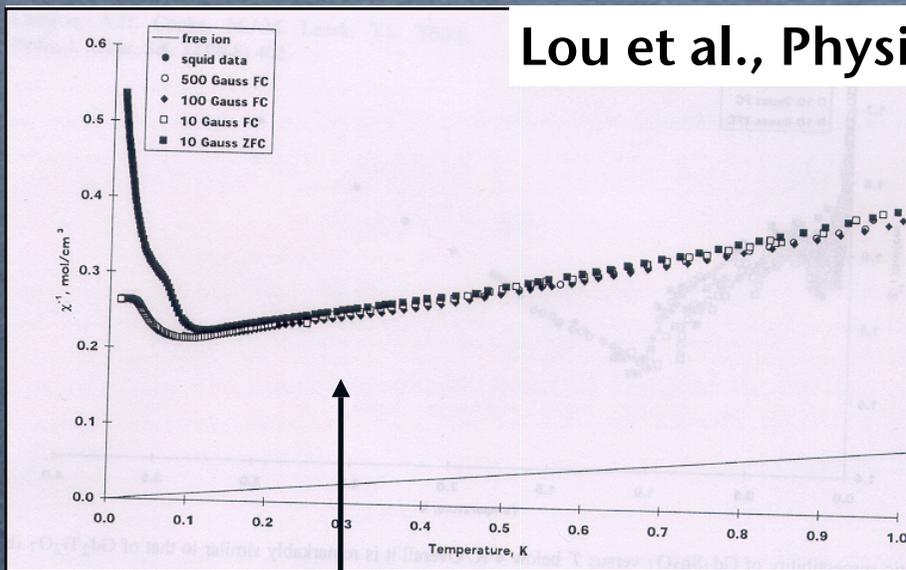


100 mK

Still Gapped

Lots of Quasielastic Scattering

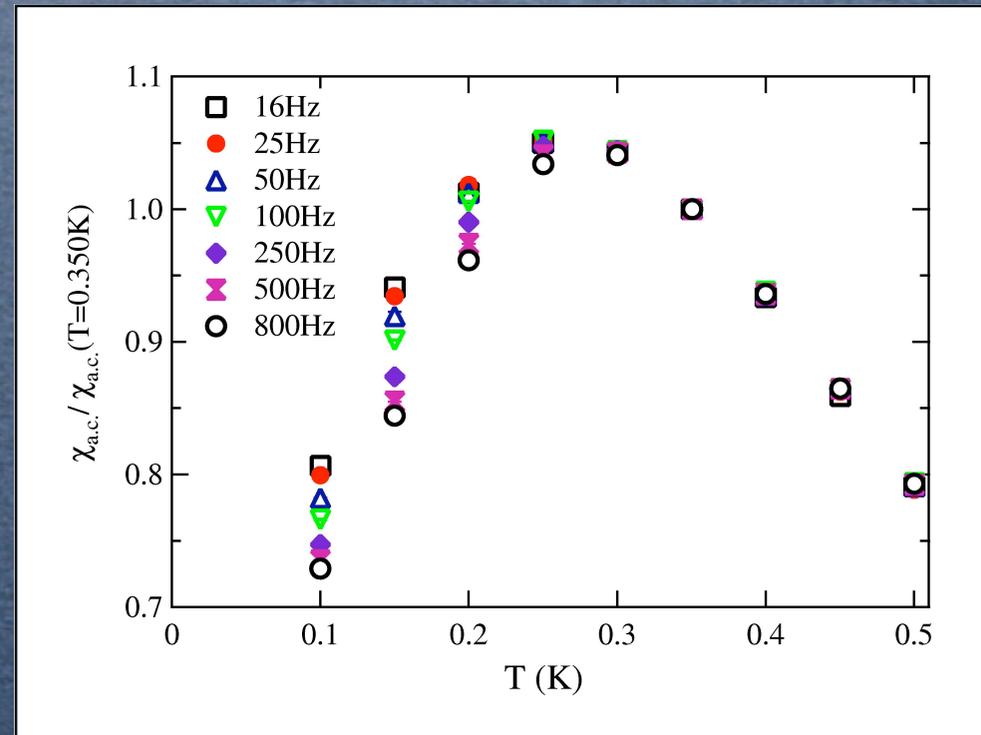
# A bit **Glassy** at low temperatures



Lou et al., Physics A 291, 306 (2001)

0.3 K

**BUT.....**



# Tb<sub>2</sub>Ti<sub>2</sub>O<sub>7</sub> - the echo

Complete relaxation at 400 mK

Baseline change at  $(0.3 \pm 0.1)$  K

**HOWEVER**

**Still** a long timescale relaxation process at 50 mK

$|Q|$  dependence

